Effect of Municipal Biowaste Biochar on Greenhouse Gas Emissions and Metal Bioaccumulation in a Slightly Acidic Clay Rice Paddy

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A field trial was performed to investigate the effect of municipal biowaste biochar (MBB) on rice and wheat growth, metal bioaccumulation, and greenhouse gas emissions in a rice paddy in eastern China. MBB was amended in 2010 before rice transplanting at rates of 0 and 40 t ha^{-1} in a field experiment lasting one cropping year. MBB soil amendment significantly increased soil pH, total soil organic carbon, and total nitrogen. The growth and grain yield of rice and wheat was not affected with MBB application at 40 t ha^{-1}. MBB amendment did not influence the soil availability of Pb, Cu, and Ni, but significantly increased the soil availability of Zn and decreased the soil availability of Cd during both rice and wheat seasons. While MBB did not change the bioaccumulation of Pb, Cu, and Ni, the rice and wheat Cd accumulation was significantly reduced, and wheat Zn accumulation slightly increased with MBB amendment. Furthermore, total N_2O emission during both rice and wheat seasons was greatly decreased, though total seasonal CH_4 emission was significantly increased in the rice season. On the other hand, soil CO_2 emission remained unaffected across crop seasons. Thus, MBB can be used in rice paddy for low carbon and low-Cd grain production, but the long-term effects remain unknown.

Keywords: Municipal biowaste; Biochar; Greenhouse gas emission; Toxic metals; Rice paddy

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INTRODUCTION

Municipal biowaste (MBW) has been considered to be a global crisis for the last several decades (Dong *et al.* 2001). In China, MBW management has been one of the major problems affecting China's environmental quality and the sustainable development of metropolitan areas (Cheng and Hu 2010). Dealing with the increasing volume of MBW presents a serious challenge for the Chinese government and scientists (Cheng and Hu 2010), as there have been serious concerns regarding the severity and rapid increase in MBW due to the fast urbanization in China (Wang and Nie 2011). The treatments for MBW in China so far include landfilling, composting, and incineration. However, there are many disadvantages to these treatments. For example, landfilling requires large areas, high capital costs, and may result in secondary pollution. Again, composting of MBW has generally shown poor quality and thus low market value of the produced compost fertilizer (Cheng and Hu 2010). In addition, MBW in China is nearly half food waste and low in calorific values (3000 to 6700 kJ/kg) but has high moisture levels (Cheng and Hu 2010; Cheng *et al.* 2007; Zhang *et al.* 2008). Consequentially, incineration of MBW entails high capital investments and especially operational costs for incineration equipment, and may increase the risk of air pollutants (Nie 2008; Cheng *et al.* 2007).

Biochar is the carbon-rich product obtained when waste biomass is pyrolysed in a closed container with restricted air conditions (Lehmann and Joseph 2009). Many biomass wastes, including crop straw, wood, bamboo, manure, and sewage sludge, are suitable feed stocks for production of biochar (Lehmann and Joseph 2009; Liu *et al.* 2013). Biochar production offers a simple, sustainable tool for waste biomass management (Lehmann and Joseph 2009). Biochar often contains a high surface area with oxygen functional groups and can absorb and hold soil nutrients (Liu *et al.* 2013; Joseph *et al.* 2013). The benefits of biochar for soil amendment include an increase in soil organic matter content, promotion of plant growth, improvement of soil structure, immobilization of soil heavy metals, reduced soil N_2O emission, and reduced nutrient leaching loss (Namgay *et al.* 2010; Cui *et al.* 2011 and 2012; Liu *et al.* 2013).

MBW in China contains a high percentage of organic matter, such as paper, food waste, wood and yard trimmings, cotton, and leather (Cheng and Hu 2010); such components offer a potential opportunity to convert MBW to biochar (MBB). Unlike many other sources of biomass, however, MBW also contains certain levels of toxic metals (Huang *et al.* 2006), which raise a critical concern for the environmental safety with application of MBB to soil. Yet, previous studies has demonstrated that the pyrolysis of sewage sludge high in heavy metals to biochar could immobilize toxic metals and thus reduce rice plant accumulation (Khan *et al.* 2013; Méndez *et al.* 2012). Whether the application of MBB will influence the accumulation of toxic metals in rice and wheat plants is still a key focus for biochar technology for MBW treatment and application in agriculture.

Atmospheric carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) are recognized as the most important long-lived greenhouse gases (GHGs). These gases significantly contribute to global warming due to their great radiative forcing (IPCC 2007a). Agricultural soils have been identified as a major source of greenhouse gas, contributing approximately 10 to 12% of the global anthropogenic GHG emissions (IPCC 2007b). Biochar soil amendment has been recommended as a key option to mitigate GHG emissions from agricultural soils (Lehmann and Joseph 2009; Sohi 2012; Khan *et al.* 2013). Previous studies have reported that biochar soil amendment could be an ecological engineering measure to mitigate global climate change by increasing soil organic carbon (SOC) storage and decreasing soil N_2O and CH_4 emissions from rice paddies in China (Zhang *et al.* 2010; Liu *et al.* 2012; Zhang *et al.* 2013). However, the effect of MBB on soil emission of greenhouse gases has not yet been addressed.

We hypothesize that MBB from MBW may affect plant growth and productivity, metal immobilization as well as reduction in greenhouse gas emission in croplands. Thus, the goal of this study is to look into the changes in productivity, plant metal bioaccumulation, and GHGs emission under MBB soil amendment, using a field experiment over a whole rice and wheat rotation in a Chinese rice paddy. We try to explore the potential of MBB to be used in rice production with regard to environmental safety and GHGs mitigation in agriculture.

EXPERIMENTAL

Materials

Biochar preparation

MBW was collected from a municipal stock of household biowaste in Shanghai Municipality, China. The raw material of MBW contained in dry base total organic carbon, N, P, and K in a range of 284.9-363.7g kg^{-1}, 6.8-8.1 g kg^{-1}, 1.6-2.0 g kg^{-1}, and 8.1-9.7 g kg^{-1}, respectively. Heavy metal contents in dry base was respectively in a range as follows: Cd in 1.4-2.4 mg kg^{-1}, Pb in 56-173 mg kg^{-1}, Cu in 36-220 mg kg^{-1}, Zn in 76-206 g kg^{-1}, and Ni in 27-43 mg kg^{-1}. Fragments of bricks, porcelains and glass were removed before treatment. The raw MBW was then dewatered by an electric extrusion press and subsequently dehydrated in the feeding chamber heated with syngas recycled from the pyrolyzer. MBW was converted to biochar *via* pyrolysis at 450 to 550 °C in a rotary kiln designed by and operated at Zhongke Anda Environment Technology Co Ltd, Shanghai, China. The detailed industrial process is presented in Supplement Fig. S1 (see Appendix). The emission of pollutants in waste gas was seen clean and could meet the standards both of China and Europe (Supplement Table S1). The product of MBB material was ground to pass through a 2-mm sieve and homogenized by thoroughly mixing before use for the experiment.

Methods

Field experiment with MBB

A field experiment with MBB amendment was carried out for a whole rice-wheat rotation year over 2010 and 2011. The site for field experiment was located in Jingtang village (31°24′N and 119°41′E), Yixing Municipality, Jiangsu Province, China. While rice has been cultivated for several thousands of years in the area, the soil was a typical high-yielding paddy soil classified as a hydroagric Stagnic Anthrosol according to Chinese Soil taxonomic Classification (Gong *et al.* 2007) and an Entic Halpudept according to Soil Taxonomy (Soil Survey Staff 1994). A subtropical monsoon climate prevailed in the area with a mean annual temperature of 15.7 °C and precipitation of 1177 mm, respectively.

MBB was added to the soil at a rate of 0 (C0 as control) and 40 t ha^{-1} (C40) after wheat harvest in May of 2010. MBB was manually spread on the soil surface and plowed to a depth of 0 to 15 cm, and homogenized subsequently with a wooden rake. No more MBB was used thereafter. Both of the control and MBB treatment were performed in triplicates, and each plot was 4 m × 4 m in area with protection rows 1 m in width. All of the plots were arranged in a randomized complete block design.

Rice seed (*Oryza sativa* L. Japonica) of the cultivar Wuyunjing 23 was directly sowed to plots on 6 June and harvested on 26 October, 2010. Similar to that reported by Zhang *et al*. (2010), rice production was managed under a typical water regime mode of M-F-D-M (moist irrigation, flooding, midseason drainage, reflooding, and moist irrigation during the seedling, tillering, stem elongation, heading and flowing, filling, and mature stages, respectively). In detail, a moist condition without submerging was kept from 5 June to 30 June; the paddy was flooded until 29 July, followed by a subsequent drainage for about 1 week before reflooding from 8 August until 30 September. A moist condition was maintained with intermittent irrigation until harvest. After rice harvest, winter wheat (*Triticum aestivum* Linn.) of the cultivar Yangfumai 4 was directly sowed on 11 November 2010 and harvested on 30 May 2011. No irrigation was performed during the wheat season.

For crop production, fertilizer was applied following the local conventional practice. For rice, total N fertilizer was applied at 300 kg N ha^{-1}, with 40% as basal fertilizer, 40% dressing at the tillering stage, and the other 20% dressing at the panicle stage. Calcium biphosphate and potassium chloride were also applied as basal fertilizer at rates of 204 kg P_2O_5 ha^{-1} and 204 kg K_2O ha^{-1} during the rice season. For wheat, the total chemical N input was 300 kg N ha^{-1} with a proportion of 60 kg N ha^{-1} as compound fertilizer ($N:P_2O_5:K_2O$=16:16:16) and the other 240 kg N ha^{-1} as urea, 30% of which was allocated as basal fertilizer, 45% as elongation fertilizer, and the other 25% as booting fertilizer.

Monitoring of greenhouse gas emissions

Monitoring of greenhouse gas emission was performed with a static chamber method following the procedure described by Zou *et al.* (2005). In each plot (Fig. 1), two aluminum flux collars (0.35 m × 0.35 m in area and 0.15 m in height) were installed after MBB amended to soil over the whole annual cycle. The top edge of each collar had a groove (5 cm in depth) for filling with water to seal the rim of the chamber with a leveled surface. For ecosystem greenhouse gas flux measurement, one collar with a chamber height of 1 m allowed crop vegetation growth with normal density. The other inter-low collar with a height of 0.25 m without covering crop vegetation allowed measurement of the soil GHG flux.

The gas was sampled in accordance with the water regime performed at different crop growth stages, which were the seedling, tillering, stem elongation, heading, flowering, grain filling, and mature stages during the rice season and the green-turning, stem elongation, heading, grain filling, and mature stages during wheat season. Gas sampling was performed between 8 and 10 a.m., and four sequential individual gas samples were collected with a syringe at 0, 10, 20, and 30 min after chamber closure. The gas sample was injected immediately after sampling into a special boron silica glass vial (No. 5, Japan Maruemu Corporation, 2010) for shipping to lab. The gas samples were analyzed with a modified gas chromatograph (Agilent 7890A) equipped with a flame ionization detector (FID) and an electron capture detector (ECD). The procedures for simultaneously measuring CO_2, CH_4, and N_2O fluxes were given in detail in a previous study (Zhang *et al.* 2010). Seasonal amounts of CO_2, CH_4, and N_2O emissions were sequentially accumulated from the emissions between every two adjacent intervals of the measurements (Zou *et al.* 2005).

Data of precipitation and temperature during the studied rice-wheat growing seasons are shown in Supplement Fig. S2. Compared to the recorded mean distribution of historical weather data, there was a 30% higher rainfall during the rice growing season and a 25% lower rainfall during the winter wheat growing season in the year of experiment (source from Yixing meteorological Bureau).

Soil and plant sampling

Soil sampling before the experiment was done after the wheat harvest in 2009 and after the rice and wheat harvests in 2010 and 2011, respectively. Composite samples from 0 to 15 cm depth were collected (Eijkelkamp soil core sampler). Soil samples were air dried and mechanically ground to < 2 mm.

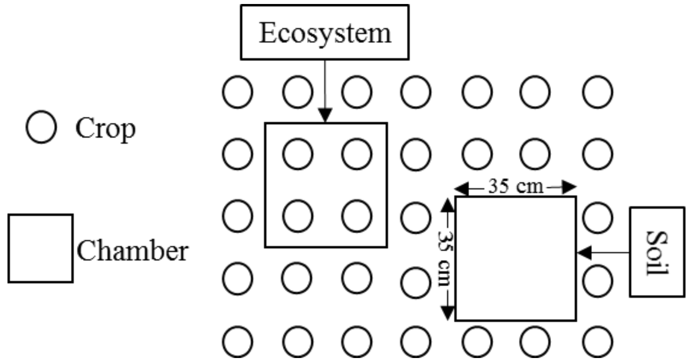

Fig. 1. Schematic diagram of chamber placement

From each plot, ten whole plant samples were collected randomly when the rice and wheat was harvested, respectively. Plant height and root length was measured after plant sampling. The samples were then washed with deionized water to remove attached soil particles. Each plant sample was then separated into roots, shoots, and grains, which were then all dried in an air-convection oven at 105 °C for 30 min, followed by 60 °C for another 48 h (Lu 2000). Unpolished rice and wheat grain samples were obtained with a thresher. The dried samples were crushed, mixed and homogenized, and stored in air-tight polyethylene bags for further chemical analysis.

Chemical analyses

Analysis of soil and MBB properties (Table 1) such as pH, cation exchange capacity (CEC), and contents of total organic carbon (TOC), total nitrogen (TN), and hot water extractable carbon (HWEC), were performed according to Zhang *et al.* (2010). Analyses of the biochar ash content and surface area were performed using a method reported by Zhang *et al.* (2011). Available P was measured using a method adapted from Rayment and Higginson (1992), while the ammonium acetate (NH_4OAc) (1 M) extraction method was used for available K (Helmke and Sparks 1996). Metal (Cd, Pb, Cu, Zn, and Ni) total concentrations in soil, MBB, and crop tissue were determined according to the protocols described by Lu (2000). In brief, soil and MBB (0.5 g) samples were digested with a mixed solution of HF-$HClO_3$-HNO_3 (10: 2.5: 2.5, v: v: v). For plant samples (0.5 g), the metals were digested with 10 mL of a mixed solution of HNO_3 and $HClO_4$ (8: 2, v: v). Digested metals were determined using graphite furnace atomic absorption spectrometry (GFAAS) (SpectrAA 220Z, Varian, USA). The DTPA (0.005 mol L^{-1}) extractable fraction of soil was measured as an assumed proxy for the bioavailable contents of metals. Surface functional groups of BC were analyzed by Fourier transform-infrared spectroscopy (FTIR, TENSOR-27, Bruker) in the range of 4000 to 400 cm^{-1} using 20 scans/min at 4 cm^{-1} resolution. Measurements were performed in pellets of biochar blended with KBr.

Calculation and statistics

Analysis of variance (ANOVA) was used to compare the differences between biochar-treated and untreated samples with the Statistical Package for Social Scientists (SPSS 16.0). In the case of significant effects, individual means were compared using the least significant difference test (LSD). A t-test was used to compare treatment effects. The significance level was defined at $p < 0.05$.

RESULTS AND DISCUSSION

Properties and Crop Growth Changes with MBB

The basic characteristics of bulk soil, MBW, and MBB before the experiment are given in Table 1. After pyrolysis, total N, P, K, and other metals were concentrated in the biochar. Compared to the bulk soil, the MBB was found to contain much higher levels of organic carbon, HWEC, N, P, K, and even heavy metals (Cd, Pb, Cu, Zn, and Ni). Due to the high pyrolysis temperature, the ash content in MBB was much higher than the wheat straw biochar (ash 20.8%) previously reported in the works by Zhang *et al.* (2013) and Bian *et al.* (2013). However, it was much similar to some other biochars reported by Chen *et al*. (2011), Bird *et al.* (2012), and Méndez *et al.* (2012). The FTIR spectra (Fig. 2) of the MBB were similar to those reported in previous studies (Cheng *et al.* 2006; Park *et al.* 2011). A broad band near 3370 cm^{-1} arising from the stretching vibration of hydroxyl groups, indicated significant hydrogen bonding interactions (Chen *et al.* 2006, 2011). The bands at 2919 and 2853 cm^{-1} were related to the elongation of CH aliphatic chains (Cheng *et al.* 2006). The bands at 2516 and 2285 cm^{-1} represented groups of S-H and of -C≡C-, respectively (Weng 2010). The small peaks at 1560 and 1800 cm^{-1} were assigned to the C=O stretching of carboxyl anions (Sun *et al.* 2013; Özçimen and Karaosmanoğlu 2004). The band at 1432 cm^{-1} was related to the deformation of CH_2 groups (Cheng *et al.* 2006). The band at 1035 cm^{-1} indicated the appearance of polysaccharide C-O (Cheng *et al.* 2006). Whereas, the bands at 877 and 817 cm^{-1} were due to the contribution from C-H bond vibration in aromatic compounds (Park *et al.* 2011).

Table 1. Basic Properties of the Studied Topsoil and MBB

Elements	Topsoil	MBB
pH (H_2O)	5.94	8.51
TOC (g kg^{-1})	26.83	310.24
Total N (g kg^{-1})	1.48	10.8
Total P (g kg^{-1})	0.34	5.26
Total K (g kg^{-1})	12.71	13.15
Available P (mg kg^{-1})	7.45	80.19
Available K (mg kg^{-1})	108.7	27.24
C/N	18.13	28.73
HWEC (mg g^{-1})	0.50	2.30
Cd (mg kg^{-1})	0.31	7.65
Pb (mg kg^{-1})	50.97	127.8
Cu (mg kg^{-1})	32.34	103.18
Zn (mg kg^{-1})	62.68	152.3
Ni (mg kg^{-1})	40.69	79.54
Surface area (m^2 g^{-1})	-	3.83
Ash (%)	-	57.99

A comparison of the properties of MBB-treated and non-treated soil is presented in Table 2. With MBB treatment, the pH of the original slightly acidic soil was significantly increased. Meanwhile, soil TOC, total N, HWEC, and available K increased for both the rice and wheat seasons. Compared to the control, the total height, root length, and grain yield of rice and wheat did not show significant changes with MBB treatment, which indicated that MBB had no significant effects on plant growth in the first year.

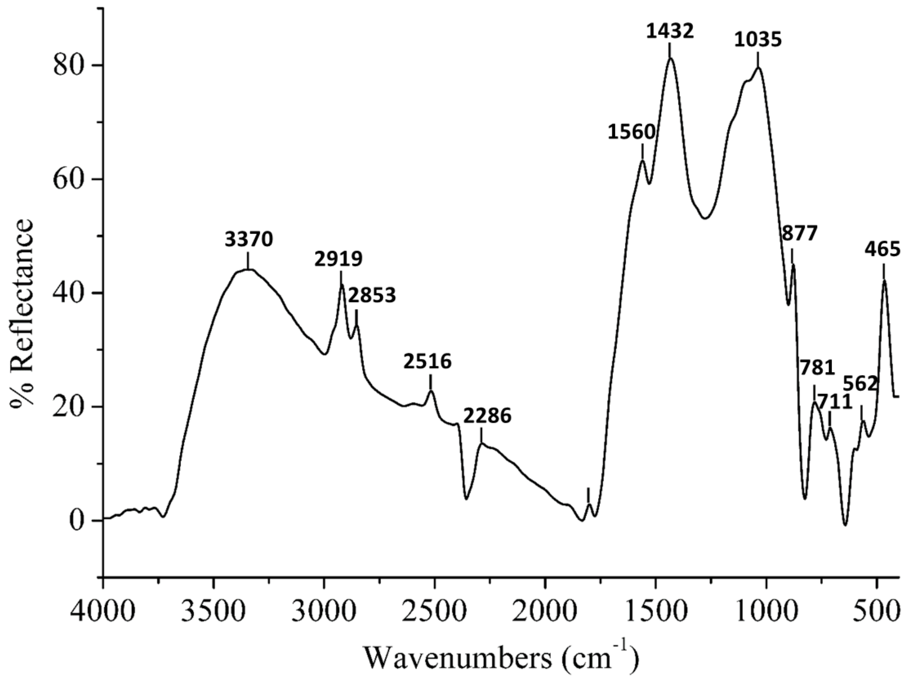

Fig. 2. FTIR spectra of municipal biowaste biochar

Table 2. Soil Properties and Crop Growth (mean ± SD, n=3) with (C40, 40 t ha-1) and without MBB (C0)

Treatment		pH (H_2O)	TOC (g kg^{-1})	TN (g kg^{-1})	Available K (mg kg^{-1})	HWEC (mg g^{-1})	Plant height (cm)	Root length (cm)	Grain Yield (t ha^{-1})
Rice	C0	5.9±0.1b	26.8±1.1b	1.48±0.02b	116±4b	0.53±0.02b	104±1a	8.6±1.2a	8.7±1.2a
	C40	6.7±0.2a	32.2±1.6a	1.58±0.02a	148±12a	0.66±0.04a	103±3a	8.5±0.9a	9.0±0.2a
Wheat	C0	5.7±0.1b	25.2±0.9b	2.21±0.09b	106±2b	0.49±0.05b	82±5a	5.7±2.0a	5.9±1.0a
	C40	6.4±0.1a	29.9±0.5a	2.37±0.03a	129±12a	0.57±0.01a	83±3a	6.1±1.9a	6.2±0.3a

Different low case letters in a single column indicate significant difference between treatments in a single crop season ($p < 0.05$)

Other studies have observed that the effect of biochar application on plant growth and productivity was highly dependent on the soil conditions, such as the pH, EC, moisture, organic matter, and fertilizers, as well as plant species (Zhang *et al.* 2012). A meta-analysis of data from over the world showed that biochar soil amendment exerted a significant yield increase of rice grain by 5.6% on average along with a liming effect (Liu *et al.* 2013). In the present study, soil pH was increased by 0.8 to 0.9, and in available K by 20% and in total N by ~10% together with an increase in SOC by 25 to 40% with the addition of MBB. But these changes did not bring about an increase in growth and productivity of rice or wheat in the soil originally rich in them. In addition, nutrient input through fertilization was already high in the present study.

Changes in Metal Bioaccumulation by Plant

The effect of MBB on the soil concentration of toxic metals is shown in Table 3. There was no significant increase in the DTPA-extractable metal pool (except Zn) under MBB treatment, while total soil concentrations of Pb, Cu, and Zn were observed significantly increased for both the rice and wheat seasons. The extent of the observed increases in total soil concentrations of Pb, Cu, and Zn were found to be between 20% and 26% and between 11% and 13% for the rice season and wheat season, respectively. However, DTPA-extractable Cd was found to be significantly decreased with MBB treatment compared to the control, by 70% and 50% for the rice and wheat seasons, respectively. However, DTPA-extractable Zn in the soil was significantly increased by 31% and 47% for the rice and wheat seasons, respectively.

As shown in Table 4, the concentrations of Cd, Pb, Cu, Zn, and Ni in rice and wheat were found in the order of root>shoot>grain, which is similar to a recent study by Khan *et al*. (2013). All the concentrations of measured metals in soil and crop grains, with MBB treatment, were below the guideline limit of the China state standard for food (GB 2762-2995), which indicated the application of MBB at 40 t ha^{-1} was safe for rice and wheat production in the study area.

Biochar soil amendment did not change the concentrations of Pb, Cu, and Ni in either rice or wheat tissues. Compared to the control, the concentration of Zn was significantly increased with MBB treatment, by 11%, 40%, and 16% for wheat grains, shoots, and roots, respectively, while a significant reduction of Cd in rice and wheat tissues was observed in this study. The average reduction rates of Cd were 50%, 41%, and 47% for rice grains, shoots, and roots, respectively. For wheat grains, shoots, and roots, the reduction rates were 30%, 25%, and 45%, respectively. Increases in crop toxic metal accumulation were often reported with application of MBW to soil (Amusan *et al.* 2005; Jiang *et al.* 2013; Chaney and Ryan 1993). However, soil amendment of MBB via pyrolysis of MBW in the present study did not cause an increase in plant accumulation of toxic metals (except Zn) of rice and wheat. This supports a safe use of MBB instead of MBW in agricultural soil, which was noted in the work Khan *et al*. (2013) with sewage sludge biochar. Though the behavior of metals in soil varies with different types of biochar treatments, the reduction in grain Cd observed here with MBB was similar to that with wheat biochar, with which the application at 40 t ha^{-1} greatly (by over 35%) reduced soil Cd mobility and grain Cd accumulation of rice across South China (Bian *et al.* (2013). Whereas, Khan *et al.* (2013) observed an increase in the bioaccumulation of Cd and Zn in rice grain grown on sewage sludge biochar-amended soil. The increase in soil pH and biochar surface functional groups may account for the reduction of soil available metals and crop metals accumulation. In the present study, the MBB used contained a much higher pH than the bulk soil (Table 1). Compared to the control, the pH of MBB-treated soil was significantly increased by 0.8 and 0.7 during the rice and wheat seasons, respectively. Bian *et al.* (2013) reported a negative correlation between soil available Cd and pH in a cross-site field experiment with wheat straw biochar treatment in China. Thus, the increase in soil pH after MBB application could immobilize soil metals to insoluble forms by precipitation or formation of hydroxide complexes, carbonates, or phosphates (Kołodyńska *et al.* 2012). Beesley and Marmioli (2011) concluded that the sorption capacity of biochar may be more important for soil metal immobilization. Uchimiya *et al.* (2011) found that the oxygen-containing carboxyl, hydroxyl, and phenolic surface functional groups of biochar play central roles in binding metal ions. Similar to these reports, MBB also contains many oxygen functional groups (Fig. 2), which can react with and immobilize toxic metals contained in soil and MBB itself.

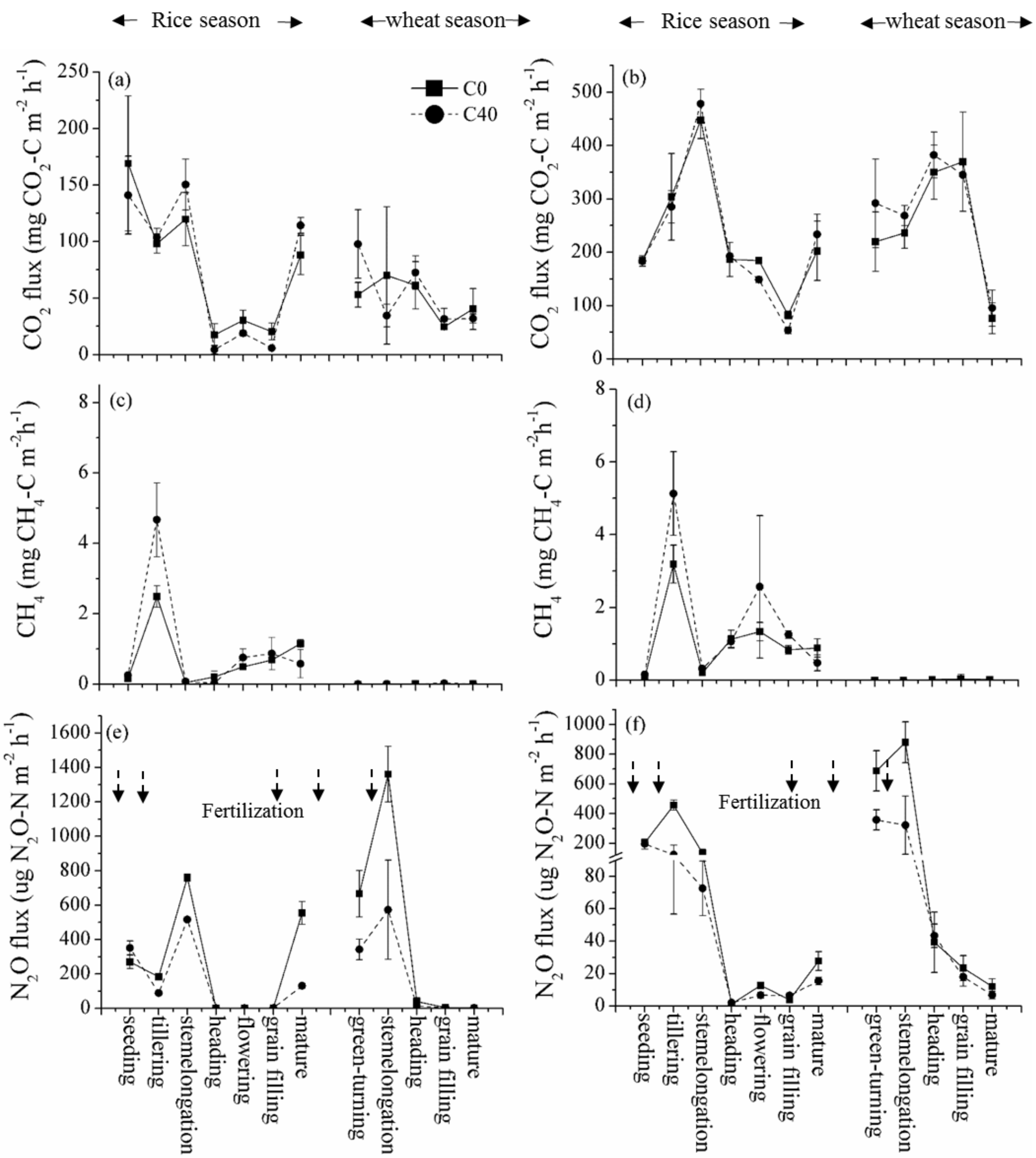

Fig. 3. CO_2, CH_4, and N_2O emissions from MBB-amended (C40) and unamend (C0) soils. (a), (c), and (e) represent emissions of CO_2, CH_4, and N_2O, respectively, from the soil; (b), (d), and (f) represent emissions of CO_2, CH_4, and N_2O, respectively, from the ecosystem

Table 3. Soil Total and DTPA Extractable Metal Concentration (mg kg^{-1}) (mean ± SD, n=3) with (C40, 40 t ha^{-1}) and without MBB (C0)

Treatment		Rice					Wheat				
		Cd	Pb	Cu	Zn	Ni	Cd	Pb	Cu	Zn	Ni
Soil total	C0	0.45±0.14a	49.6±3.0b	31.5±1.0b	61.1±1.7b	39.7±1.6a	0.47±0.17a	53.8±2.1b	31.1±0.8b	58.3±2.4b	34.6±1.0a
	C40	0.54±0.31a	59.3±9.0a	39.7±7.4a	75.5±6.7a	39.8±1.9a	0.57±0.12a	63.4±5.4a	34.9±2.5a	65.9±1.7a	33.3±2.4a
DTPA-extractable	C0	0.10±0.01a	8.8±1.0a	6.6±0.5a	5.7±0.5b	1.87±0.2a	0.16±0.00a	12.6±0.9a	8.5±0.8b	5.8±0.4b	2.3±0.2a
	C40	0.03±0.01b	10.1±1.7a	7.7±0.6a	7.5±0.3a	1.63±0.2a	0.08±0.00b	13.3±2.2a	9.0±0.4b	8.6±0.3a	1.9±0.1a

Different letters in a single column indicate significant difference between treatments in a single crop season ($p < 0.05$)

Table 4. Metal Concentration (mg kg^{-1}) (mean ± SD, n=3) of Crop Grains, Shoots, and Roots with (C40, 40 t ha^{-1}) and without MBB (C0)

Treatment		Rice					Wheat				
		Cd	Pb	Cu	Zn	Ni	Cd	Pb	Cu	Zn	Ni
Grain	C0	0.06±0.04a	0.04±0.02a	1.16±0.54a	15.54±4.47a	1.81±0.63a	0.23±0.03a	0.05±0.03a	6.96±1.05a	41.11±2.12b	0.76±0.48a
	C40	0.03±0.01b	0.08±0.04a	1.33±0.15a	16.89±3.08a	1.76±0.33a	0.16±0.02b	0.04±0.02a	6.98±0.62a	45.56±3.24a	0.67±0.21a
Shoot	C0	0.34±0.04a	1.27±0.44a	6.37±0.88a	39.06±1.43a	3.61±1.30a	0.28±0.05a	0.35±0.01a	3.90±1.14a	22.46±3.38b	1.08±0.55a
	C40	0.20±0.03b	1.34±0.21a	6.51±0.74a	39.17±0.78a	3.57±1.22a	0.21±0.01b	0.42±0.11a	4.68±1.34a	31.44±5.50a	1.19±0.15a
Root	C0	2.92±0.37a	24.24±5.23a	40.46±3.01b	51.40±1.22b	55.99±17.27a	0.77±0.12a	1.11±0.42a	13.58±3.02b	61.41±1.62b	1.05±0.23a
	C40	1.54±0.24b	30.43±7.09a	35.19±6.36a	55.69±2.00a	36.44±4.16a	0.42±0.06b	1.34±0.08a	19.36±3.39a	71.48±4.49a	0.97±0.14a

Different letters in a single column indicate significant difference between treatments in a single season ($p < 0.05$)

Meanwhile, a previous study showed that the MBB used in this study could adsorb Cd^{2+} in solution and that the maximum adsorption capacity was 6.22 mg g^{-1}, as determined using the Langmuir model (Qin *et al.* 2012); this may explain the reduction in bioavailable soil Cd with MBB treatment. The increase in soil available Zn in the present study could be due either to (1) a high initial Zn content in soil and MBB (Table 1), or (2) a high desorption efficiency of Zn on the biochar surface caused by other competitive metals (Cu, Pb, and Cd) (Trakal *et al.* 2011).

Effect of MBB on GHG_S Emissions

The changes in soil and ecosystem emissions of CO_2, CH_4, and N_2O with MBB treatment are presented in Fig. 3. The temporal distribution pattern of soil and ecosystem CO_2 fluxes over the whole crop growing season were independent of biochar treatment (Figs. 3 a, b). Clearly, there was no significant increase in the total emission of CO_2 with MBB treatment during both the rice and wheat seasons (Table 5). In contrast to the findings with laboratory incubation study by Zimmerman (2010), the results here indicated an absence of the priming effect of biochar soil amendment which had been argued in previous studies. The high recalcitrance of organic carbon content in biochar could be responsible for the insignificant change in CO_2 emission with MBB addition (Jones *et al.* 2011). This had been often addressed as biochar stability (Lehmann & Sohi 2008; Wardle *et al.* 2008).

Table 5. Total CO_2, CH_4 and N_2O Emissions (mean ± SD, n=3) from the Paddy under Rice-Winter Wheat Cropping with (C40, 40 t ha^{-1}) and without (C0) MBB

Treatment		Rice season			Wheat season	
		CO_2 (kg CO_2-C ha^{-1})	CH_4 (kg CH_4-C ha^{-1})	N_2O (kg N_2O-N ha^{-1})	CO_2 (kg CO_2-C ha^{-1})	N_2O (kg N_2O-N ha^{-1})
Soil	C0	2042±215a	24.8±2.5b	7.4±0.3a	983±309a	10.5±1.4a
	C40	2146±76a	35.9±3.4a	3.7±0.2b	1141±169a	4.7±1.7b
Eco-system	C0	6827±783a	37.5±5.2b	3.6±0.1a	4808±215a	7.9±0.3a
	C40	6824±131a	45.5±6.2a	1.4±0.3b	5330±500a	3.4±0.9b

Different letters in a single column indicate significant difference between treatments in a single crop season ($p < 0.05$)

In the present study, CH_4 emission was mainly observed during the rice season, while it was in trace amounts and even negative during the wheat season (Fig. 3 c, d). All the fluxes were greater from ecosystem than from soil as CH_4 could be largely emitted due to transport from rhizosphere via the intercellular plant-mediated (Schütz *et al.* 1989). MBB soil amendment did not change the dynamic pattern of CH_4 emission during the rice and wheat seasons. Emission of rice ecosystem CH_4 peaked both at the tillering and flowering stages during flooding or irrigation. For rice soil, however, the peak of CH_4 emission was found only under flooding during the tillering stage while flooded. Compared to the control, the rice ecosystem CH_4 emissions at the tillering and grain-filling stages significantly increased with MBB treatment, by 61% and 93%, respectively. During the rice season, the flux of soil CH_4 significantly increased (by 88%) in the tillering stage, but significantly decreased in the mature stage (50%). Compared to the control, total rice seasonal total CH_4 emissions significantly increased, by 45% and 21% for the soil and ecosystem, respectively (Table 5). Khan *et al.* 2013 reported that biochar soil amendment could decrease paddy soil CH_4 emission by increase soil aeration. But the result of this study was in good agreement with our previous study. Zhang *et al.* (2011) reported that wheat straw biochar applied (40 t ha^{-1}) in a rice paddy, adjacent to the field of this study, significantly increased soil total CH_4 emission by 49%, compared to the control. This was attributed to the existence of the soluble organic carbon shortly after the biochar amendment, as rates of CH_4 production could be primarily determined by the availability of carbon substrate for methanogens other than the impact of environmental variables (Van Denier der Gon & Neue 1995). Knoblauch *et al.* (2010) argued that the labile organic C pool of biochar could be decomposed and become the predominant source of methanogenic substrates, thus promoting CH_4 production, particularly in the early stages of rice production. A relatively higher HWEC in MBB and MBB-treated soil (Tables 1 and 2) may partly explain the increase in soil CH_4 emission in this study. On the other hand, a high rate of biochar amendment of 40 t ha^{-1} induced a high C/N ratio, which could give rise to an N limitation to methanogens bacteria. Of course, the mechanism behind this phenomenon is still unclear and need further study.

Similar to CO_2 and CH_4 emissions, the seasonal dynamics of N_2O flux were not affected by MBB amendment but largely dependent on fertilizer application and water regime (Fig. 3 e and f). Soil N_2O emission peaked in the seeding and tillering stages for the rice season and in the green-turning and stem elongation stages for the wheat season, respectively. For the ecosystem, N_2O peaked in the stem elongation stage for rice season and in the green-turning and stem elongation stages for wheat season, respectively. MBB soil amendment significantly decreased both the soil and ecosystem N_2O emission, especially during the growing stages with high N_2O flux. Several reasons can be given to explain these N_2O dynamics.

Biochar in soil might act as an “electron shuttle” to facilitate the activity of N_2O reductase from denitrifying microorganisms as soil pH was increased (Yanai *et al.* 2007) and therefore promote the last step of denitrification (Cayuela *et al.* 2013). Biochar can adsorb anions such as NO^{3-} (Mizuta *et al.* 2004; Novak *et al.* 2010; Cayuela *et al.* 2013), which may lead to a decrease in the denitrification N_2O (Baggs *et al.* 2000). Huang *et al.* (2004) found that the cumulative emission of N_2O was negatively correlated with the C/N ratio of soil amendment.

As reported in previous studies, significant effects were shown with biochar amendment on N_2O emissions from croplands (Zhang *et al.* 2010; Liu *et al.* 2012; Taghizadeh-Toosi *et al.* 2011). Here again, the total soil N_2O emission significantly decreased with MBB treatment, by 50% and 56% during the rice and wheat seasons, respectively (Table 5). For the ecosystem, the reductions in the total N_2O emissions during the rice and wheat seasons were 60% and 57%, respectively. Wheat straw biochar applied at 40 t ha^{-1} to an adjacent paddy in the work by Zhang *et al.* (2013) also reported the total ecosystem N_2O emission was significantly reduced by 39% for the rice season and 41% for the wheat season. The findings in the present study may confirm the positive effect of biochar on reducing rice paddy N_2O emission.

CONCLUSIONS

1. This study envisaged a new option for municipal bio-waste treatment. Conversion of municipal bio-waste to biochar and its application at 40 t ha^{-1} to paddy soil greatly decreased soil and ecosystem greenhouse gas emissions, while no negative effect was observed for rice and wheat growth or grain yield.
2. Municipal biochar application to paddy soil significantly reduced soil DTPA-extractable Cd and rice and wheat Cd accumulation but no statistical increase in plant accumulation of Pb, Cu, and Ni though these metals were present in the biochar.
3. The long-term changes in the effect of municipal biowaste biochar on soil metal mobility, greenhouse gas emission, and plant growth, as well as the biochar-soil interactions remain unclear.

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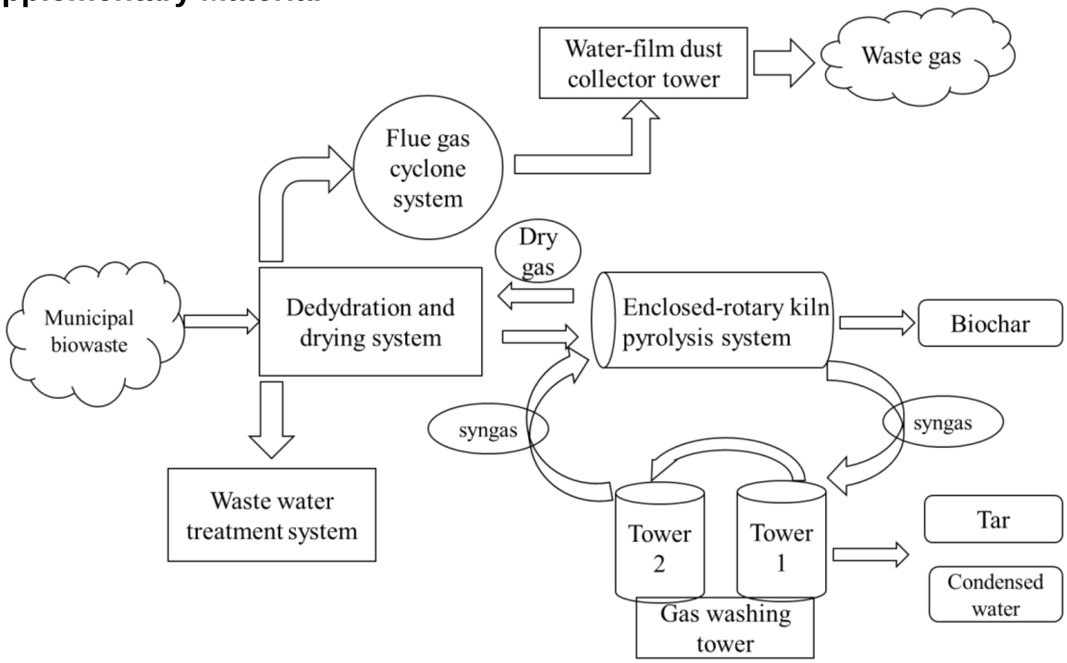
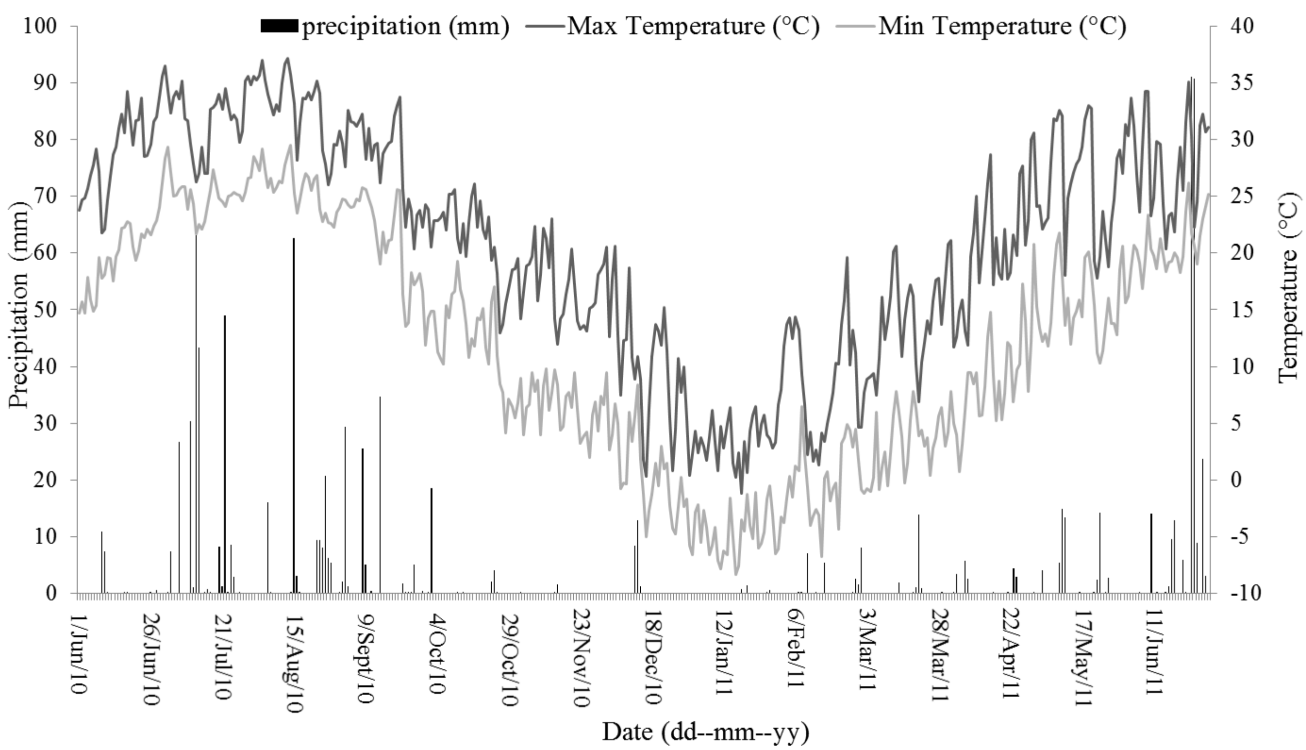
APPENDIX

Supplementary Material



Supplement Fig. S1. Schematic diagram of the MSW pyrolysis facility



Supplement Fig. S2. Daily precipitation (vertical bars) and maximum (dark curve) and minimum (gray curve) air temperature over the rice-wheat rotation cycle studied

Supplement Table S1. Pollutants Emission from Pyrolyzer in this Study and Comparison with Maximum Permissible Limits Set for Municipal Biowaste Incineration in China and Europe

Pollutants	Pyrolyzer emission	GB 18485-2001[a]	EU-2000/76/EC[b]
Particulate matter(mg m^{-3})	15.73	80	30
CO (mg m^{-3})	82.27	150	50
NO_X (mg m^{-3})	13.33	400	500
SO_X (mg m^{-3})	ND	260	50
HCl (mg m^{-3})	3.26	75	10
Hg (mg m^{-3})	1.21×10^{-3}	0.2	0.05
Cd (mg m^{-3})	ND	0.1	0.05
Pb (mg m^{-3})	4.77×10^{-3}	1.6	0.5
Dioxins (ng-TEQ Nm^{-3})	2.76×10^{-5}	1	0.1

[a]Standard for pollution control on municipal solid waste incineration in China
[b]Directive 2000/76/EC on the incineration of waste, Official Journal of the European Communities