Biochar can decrease the gaseous reactive nitrogen intensity in intensive vegetable soils across mainland China

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Highlights

1. Two contrasting biochars affected GNrI across 4 major vegetable soils in China.
2. Biochar affects gaseous Nr or yield largely depending on soil types.
3. Both biochars decreased GNrI with Bw mitigated gaseous Nr whereas Bm improved yield.

Abstract

Biochar amendment to soil has been proposed as a strategy for sequestering carbon, mitigating climate change and enhancing crop productivity, but few studies have demonstrated the general effects of different feedstock-derived biochars on the various gaseous reactive nitrogen emissions (GNrEs, N_2O, NO and NH_3) simultaneously across the typical vegetable soils in China. A greenhouse pot experiment with five consecutive vegetable crops was conducted to investigate the effects of two contrasting biochar, namely, wheat straw biochar (Bw) and swine manure biochar (Bm) on GNrEs, vegetable yield and gaseous reactive nitrogen intensity (GNrI) in four typical vegetable soils from Hunan province (HN), Shanxi province (SX), Shandong province (SD) and Heilongjiang province (HLJ) which are representative of the intensive vegetable ecosystems across mainland China. Results showed that remarkable GNrE mitigation induced by biochar occurred in SX and HLJ soils, whereas enhancement of yield occurred in SD and HLJ soils. Additionally, both biochars decreased GNrI with Bw performed better than Bm regarding N_2O mitigation, with Bw mitigating N_2O and NO emissions by 21.8–59.1 % and 37.0–49.5 % (except for SD), respectively, while Bm improved yield by 13.5–30.5 % (except for HN and SX).Biochar amendments generally stimulated the NH_3 emissions with greater enhancement from Bm than Bw. We can infer that the biochar's effects on the GNrEs and vegetable yield strongly depend on the attributes of the soil and biochar. Therefore, both soil type and biochar characteristics should be seriously considered before conducting large-scale application of biochar in order to achieve the maximum benefits under intensive greenhouse vegetable agriculture.

Keyword: Biochar, Intensive vegetable soil, Gaseous reactive nitrogen emissions (GNrEs), Gaseous reactive nitrogen intensity (GNrI)

1 Introduction

Agriculture accounted for an estimated emission of 4.1 (1.7–4.8) Tg N yr^{-1} for N_2O and 3.7 Tg N yr^{-1} for NO, contributing 60 % and 10 %, respectively, to the total global anthropogenic emissions, largely due to increases of N fertilizer application in cropland (Ciais, 2013). The concentration of atmospheric N_2O, a powerful, long-lived, greenhouse gas, has increased from 270 parts per billion by volume (ppbv) in the pre-industrial era to ~ 324 ppbv (Ussiri and Lal, 2013); it has 265 times the global warming potential (GWP) of CO_2 on a 100-year horizon (IPCC, 2013) and also causes depletion of the ozone layer in the atmosphere (Ravishankara et al., 2009). In contrast, NO_x, which is mainly emitted as nitric oxide (NO), does not directly affect the earth's radiative balance but catalyzes the production of tropospheric ozone (O_3), which is a greenhouse gas associated with detrimental effects on human health (Anenberg et al., 2012) and crop production (Avnery et al., 2011). Additionally, along with the high nitrogen (N) application, ammonia volatilization is one of the major N loss pathways (Harrison and Webb, 2001) as well, with up to 90% coming from agricultural activities (Misselbrook et al., 2000; Boyer et al., 2002). As a natural component and a dominant atmospheric alkaline gas, NH_3 plays an important role in atmospheric chemistry and ambient aerosol formation (Langridge et al., 2012; Wang et al., 2015b). In addition to nutrient enrichment (eutrophication) of terrestrial and aquatic systems and global acidification of precipitation, NH_3 has also been shown to be a major factor in the formation of atmospheric particulate matter and secondary aerosols (Kim et al., 2006; Pinder et al., 2007), leading to potentially adverse effects on human and ecosystem health such as visibility degradation and threats to biodiversity (Powlson et al., 2008; Behera et al., 2013). Consequently, the release of various reactive N species results in lower N use efficiency in agricultural systems.

In China, vegetable production devotes an area of approximately 24.7 $\times 10^6$ ha, equivalent to 12.4% of the total available cropping area, and the production represented 52 % of the world vegetable production in 2012 (FAO, 2015). Intensified vegetable cultivation in China is characterized by high N application rates, high cropping index and frequent farm practices. Annual nitrogen fertilizer inputs for intensively managed vegetable cultivation in rapidly developing areas are 3–6 times higher than in cereal grain cultivation in China (Ju et al., 2006; Diao et al., 2013; Wang et al., 2015a). As a result, great concern exists about excess N fertilizer application, leading to low use efficiency in intensive vegetable fields in China (Deng et al., 2013; Diao et al., 2013). Meanwhile, intensive vegetable agriculture is considered to be an important source of N_2O (Xiong et al., 2006; Jia et al., 2012; Li et al., 2015b; Zhang et al., 2015) and NO production (Mei et al., 2009). Moreover, ammonia volatilization is another important N pathway in fertilized soil, resulting in large losses of soil-plant N (Pacholski et al., 2008; Zhang et al., 2011). Therefore, the reduction of reactive N loss becomes a central environmental challenge to meet the joint challenges of high production and acceptable environmental consequences in intensive vegetable production (Zhang et al., 2013).

Biochar is the dark-colored, carbon (C)-rich residue of pyrolysis or gasification of plant biomass under oxygen (O_2)-limited conditions, specifically produced for use as a soil amendment (Sohi, 2012). The amendment of agricultural ecosystems with biochar has been proposed as an effective countermeasure for climate change (Smith, 2016). These additions would increase soil carbon storage (Mukherjee and Zimmerman, 2013; Stavi and Lal, 2013), decrease GHG emissions (Li et al., 2016), and improve soil fertility and crop production (Major et al., 2010; Liu et al., 2013). However, some recent studies have reported no difference or even an increase in soil N_2O emissions induced by biochar application from different soils (Saarnio et al., 2013; Wang et al., 2015a). Still, NH_3 volatilization was enhanced by biochar application in pasture soil (Clough et al., 2010), vegetable soil (Sun et al., 2014) and paddy soil in the wheat-growing season (Zhao et al., 2014). Additionally, crop productivity responses to biochar amendments differed among various biochars (Cayuela et al., 2014). These inconsistent results suggest that current biochar application to soil is not a "one-size fit-all paradigm" because of the variation in the physical and chemical characteristics of the different biochars, soil types and crop species (Field et al., 2013; Cayuela et al., 2014). Moreover, limited types of biochar (Spokas and Reicosky, 2009) and soil (Sun et al., 2014) were involved in the experiments in previous studies. Thus, the evaluation of the different types of biochar under the typical soils is imperative to gain a comprehensive understanding of potential interactions before the large-scale application of biochars in intensive vegetable cropping system in China.

Therefore, a greenhouse pot experiment was conducted in an effort to investigate the effects of different types of biochar on gaseous reactive nitrogen emissions (GNrEs), namely, N_2O, NO and NH_3, simultaneously in four typical intensified vegetable soils across main vegetable production areas of mainland China. Overall, the objectives of this research were to gain a comprehensive insight into the effects of the different types of biochar on the GNrEs, vegetable yield and gaseous reactive nitrogen intensity (GNrI) in intensively managed vegetable production in China.

2 Materials and methods

2.1. Experimental soil and biochar

Four typical greenhouse vegetable cultivation sites with a long history (more than 10 years) of conventional cultivation were selected from Northeast, Northwest, Central and Eastern China (Fig. S1), namely, Phaeozem, Anthrosol, Acrisol and Cambisol (FAO and ISRIC, 2012) from Jiamusi (46°48´N, 130°12´E), Heilongjiang province (HLJ); Yangling (34°18´N, 108°2´E), Shanxi province (SX); Changsha (28°32´N, 113°23´E), Hunan province (HN) and Shouguang (36°56´N, 118°38´E), Shandong province (SD), respectively were collected and represented a range of differences in physicochemical properties and regions (Table S1). Soil samples were manually collected from the cultivated layer (0–20 cm) after the local vegetable harvest in April, 2015. The samples were air-dried and passed through a 5 mm stainless steel mesh sieve and homogenized thoroughly. Any visible roots and organic residues were removed manually before being packed with the necessary amount of soil to achieve the initial field bulk density. Each pot received 15 kg of 105 ℃ dry-weight-equivalent fresh soil. For each of the biochar amendment pot, 282.6 g pot^{-1} sieved biochar (2 mm) was mixed with the soil thoroughly before the experiment, which was equivalent to a 40 t ha^{-1} biochar dose (dry weight). No more biochar was added later in the experimental period.

Two types of biochar, derived from two common agricultural wastes in China: wheat straw and swine manure, hereafter referred to as Bw and Bm, respectively (Table S1). The Bw was produced at the Sanli New Energy Company in Henan, China, by pyrolysis and thermal decomposition at 400–500 ℃. The Bm was produced through thermal decomposition at 400 ℃ by the State Key Laboratory of Soil Science and Sustainable Agricultural, Institute of Soil Science, Chinese Academy of Sciences. In accordance with Lu (2000), the SOC was measured by wet digestion with H_2SO_4–$K_2Cr_2O_7$, TN was determined by semi-micro Kjeldahl digestion, and soil texture was determined with the pipette method. The soil pH and biochar pH were measured in deionized water at a volume ratio of 1:2.5 (soil to water) with a PHS-3C mv/pH detector (Shanghai Kangyi Inc. China). Biochar content of hydrogen (H) was measured by elemental analysis after dry combustion (Euro EA, Hekatech GmbH, Wegberg, Germany). The oxygen content of biochar was measured with the same device after pyrolysis of the sample at 1000 ℃ followed by reduction of the evolved O_2 to CO and quantification by GC-TCD. The soil NO_3^-–N and NH_4^+–N were measured following the two-wavelength ultraviolet spectrometry and indophenol blue methods, respectively, using an ultraviolet spectrophotometer (HITACHI, UV-2900, Tokyo, Japan). Electric conductivity (EC) was measured by using a Mettler-Toledo instrument (FE30-K, Shanghai, China) at a 1:5 (w:v) soil to water ratio. Cation exchange capacity (CEC) was determined using the CH_3COONH_4 method. Dissolved organic carbon (DOC) was extracted from 5 g of the biochar/soil with an addition of 50 ml deionized water and measured by a TOC analyzer (TOC-2000/3000, Metash Instruments Co., LTD, Shanghai, China). Ash content was measured by heating the biochars at 750 ℃ for 4 h. The specific surface area of the biochar material was tested using the Brunauer–Emmett–Teller (BET) method, from which the N adsorption–desorption isotherms at 77 K were measured by an automated gas adsorption analyzer ASAP2000 (Micromeritics, Norcross, GA) with + 5% accuracy. Scanning electron microscopy (SEM) imaging analysis was conducted using a HITACHI S-3000N scanning electron microscope.

2.2. Experimental set-up and management

The pot experiments were performed at the greenhouse experimental station of Nanjing Agricultural University, China. Five vegetable crops were grown successively in the four vegetable soils during the experimental period. For each type of soil, three treatments with three replicates were arranged in a random design: urea without biochar (N), urea with wheat straw biochar (N+Bw), urea with swine manure biochar (N+Bm). In addition, phosphate and potassium fertilizers in the form of calcium magnesium phosphate and potassium chloride, together with urea, were broadcasted and mixed with soil thoroughly prior to sowing the vegetables. No topdressing events occurred because of the frequent cultivation and short growth period for the leafy vegetables. Based on the vegetable growth, all pots received equal amounts of water and no precipitation. Detailed information on the pot management practices is provided in Table S2.

Each pot consists of a 30 cm × 30 cm (height × diameter) cylinder made of polyvinyl chloride (PVC). The top of each pot was surrounded by a special water-filled trough collar, which allowed a chamber to sit on the pot and prevent gas exchange during the gas-sampling period. Small holes (diameter of 1 cm) at the bottom of the pots were designed for drainage. To prevent soil loss, a fine nylon mesh (< 0.5 mm) was attached to the base of the soil cores before packing.

2.3. Measurement of N_2O, NO and NH_3

The NO and N_2O fluxes were measured simultaneously from each vegetable cultivation using a static opaque chamber method (Zheng et al., 2008; Yao et al., 2009). A square PVC chamber of 35 cm × 35 cm × 40 cm (length × width × height) was temporarily mounted on the pot for gas flux measurement. The chamber was coated with sponge and aluminum foil outside to prevent solar radiation heating the chamber. Gas samples for flux measurements were collected between 8 and 10 a.m. on each measuring day to minimize the influence of diurnal temperature variation. Gas fluxes were usually measured once a week and every other day for one week following fertilizer application. To measure the N_2O flux, four samples were collected from the headspace chamber using 20 ml polypropylene syringes at 0, 10, 20, and 30 min after chamber closure. The gas concentrations in the samples were analyzed within 12 h after sampling using an Agilent 7890A gas chromatograph equipped with an electron capture detector (ECD) for N_2O detection. The carrier gas was argon-methane (50 %) at a flow rate of 40 ml min^{-1}. The column and ECD temperatures were maintained at 40 and 300 ℃, respectively. The gas chromatography configurations described by Wang et al. (2013) were adopted for the gas concentration analysis. N_2O flux was calculated using the linear increases in gas concentration with time. Sample sets were rejected unless they yielded a linear regression value of $R^2 > 0.90$.

For each NO flux measurement, gas samples were collected from the same chamber that was used for the N_2O flux measurements (Yao et al., 2009). Before closing the chamber, an approximately 1.0 L gas sample from the headspace of each chamber was extracted into an evacuated sampling bag (Delin Gas Packing Co., LTD, Dalian, China), and this measurement was regarded as time 0 min for NO analysis. After 30 min under chamber enclosure conditions (i.e., after the N_2O sample collections were completed), another headspace gas sample with the same volume was extracted from each chamber into another evacuated bag. Within 1 h after sampling, NO concentrations were analyzed by a model 42*i* chemiluminescence NO–NO–NO_X analyzer (Thermo Environmental Instruments Inc., Franklin, MA, USA). The NO fluxes were derived from the concentration differences between the two collected samples. The NOx analyzer was calibrated by a model 146*i* dynamic dilution calibrator system at the end of each crop-growing season.

The mean flux of N_2O or NO during the experiment period was calculated as the average of all measured fluxes, which were weighted by the interval between the two measurements (Xiong et al., 2006). The cumulative N_2O was calculated as the product of the mean flux and the entire duration.

The NH_3 volatilization was determined using the ventilation method (Zhao et al., 2010). The phosphoglycerol-soaked sponge was replaced every day after each fertilization event for approximately one week. The phosphoglycerol-soaked sponges used to collect the NH_3 samples were immediately extracted with 300 mL potassium chloride (KCl) solution (1 mol L^{-1}) for 1 h. The concentration of ammonia nitrogen (NH_4^+–N) was measured using the indophenol blue method at 625 nm (Sororzano, 1969) by ultraviolet spectrophotometry (HITACHI, UV-2900, Tokyo, Japan, with 0.005 absorbance of photometric accuracy). The cumulative seasonal NH_3 volatilization was the sum of the daily emissions during the measurement period.

2.4. Auxiliary measurements

Simultaneously with the determination of trace gas fluxes, the air temperature and the soil temperature at a depth of 5 cm were measured using thermally sensitive probes at each sampling date. Soil water content was also measured using a portable water detector (Mode TZS-1K, Zhejiang Top Instrument Corporation Ltd., China) by the frequency domain reflectometer method at a depth of 5 cm. Measured soil water contents (v/v) were converted to water filled pore space (WFPS) with the following equation:

$$\text{WFPS} = \text{volumetric water content } (\text{cm}^3\ \text{cm}^{-3}) \,/\, \text{total soil porosity } (\text{cm}^3\ \text{cm}^{-3}) \qquad (1)$$

Here, total soil porosity = [1 − (soil bulk density (g cm^{-3}) / 2.65)] with an assumed soil particle density of 2.65 (g cm^{-3}). The total soil bulk density was determined with the cutting ring method according to Lu (2000).

After each vegetable crop reached physiological maturity, the fresh vegetable yield was measured by weighing the whole aboveground and belowground biomass in each pot.

$$GNrE = \text{cumulative } N_2O + \text{cumulative NO} + \text{cumulative } NH_3 \text{ emissions (kg N ha}^{-1}) \quad (2)$$

$$GNrI = GNrE / \text{vegetable fresh yield (kg N t}^{-1} \text{ yield)} \quad (3)$$

After the one-year pot experiment, a soil sample from each pot was blended carefully. One subsample was stored at 4 ℃ for determination of microbial biomass carbon (MBC), potential nitrification rate (PNR) and denitrification enzyme activity (DEA) within 3 days. Another subsample was air-dried for analysis of SOC, TN, pH and EC. MBC was determined by substrate-induced respiration using a gas chromatography (Anderson and Domsch 1978). PNR was measured using the chlorate inhibition soil-slurry method as previously described (Kurola et al., 2005) with modifications (Hu et al., 2016). DEA was quantified as described by Smith and Tiedje (1979).

2.5. Data processing and statistics

One-way ANOVA was performed to test the effects of the treatments on cumulative N_2O, NO and NH_3 emissions; GNrE; vegetable yield and GNrI. Two-way ANOVA was used to analyze the effects of the biochar type; soil type; and their interactions on N_2O, NO and NH_3 emissions, vegetable yield, GNrE and GNrI throughout the experimental period. Multiple comparisons among the treatments were further explained using Tukey's HSD test. Significant differences were considered at $P < 0.05$. All statistical analyses were performed using JMP ver. 7.0 (SAS Institute, Cary, NC, USA, 2007). Pearson's correlation analysis was used to determine whether there were significant interrelationships between N_2O/NO and PNR or DEA in each soil, using SPSS window version 18.0 (SPSS Inc., Chicago, USA).

3. Results

3.1. Soil responses to biochar amendment

Obvious differences in all observed soil properties existed among soil types (Table 1, $p < 0.001$), suggesting the wide variations of soil characters across mainland China. Additionally, biochar amendments had significant influences on all the soil properties (Table 1, $p < 0.05$). Compared with N treatments, biochar amendments increased the SOC, TN and EC by 20.4–135.0 %, 0.5–21.2 % and 2.4–38.1 %, respectively, across all the soils. Compared with Bw, Bm amendment resulted in higher contents of SOC and TN by 5.8–20.5 % and 9.5–14.2 %, respectively, whereas EC values were higher by 3.3–21.5 % induced by Bw than Bm amendment over all soils. Additionally, biochar amendments significantly increased soil pH by 0.27–0.64 and 0.08–0.10 units compared with N treatment in HN and SX soils ($p < 0.05$), respectively, and higher values were detected with Bm than Bw amendment in all soils. Furthermore, biochar amendments tended to increase MBC in SD and HLJ soils, and Bm performed better in MBC enhancements than Bw in all soils.

As shown in Fig. 1, no consensus effects on PNR and DEA were observed with biochar amendments across all soils. Compared with N treatment, biochar amendments significantly increased PNR in HLJ while exerted no influences on SD soil (Fig. 1a). Compared with Bw, Bm amendment significantly increased PNR in HN and SX soils. Moreover, compared with N, biochar amendments reduced DEA in most soils, significantly in SX and HLJ by an average of 40.1 and 37.8 % (Fig. 1b, $p < 0.05$), respectively. In comparison with Bw, remarkable enhancements in DEA were observed by 42.5 and 74.4 % with Bm amendment in HN and SX soils, respectively ($p < 0.05$).

3.2. Seasonal variations of N_2O and NO emissions

The dynamics of N_2O fluxes from all N-applied treatments in the four vegetable soils were relatively consistent and followed a sporadic and pulse-like pattern that was accompanied with fertilization, tillage and irrigation (Fig. 2). In addition, peak N_2O fluxes varied greatly. Most of the N_2O emissions occurred during the Amaranth and Tung choy growing periods, and there were several small emissions peaks during the Spinach and Coriander herb growing periods due to lower N application rate (Table S2), soil temperature and water content (Fig. S2). The highest peaks of N_2O emissions from HN, SX, SD and HLJ were 4133.7, 1784.0, 432.4 and 1777.2 μg N m^{-2} h^{-1}, respectively. Although biochar (Bw and Bm) application did not significantly alter the seasonal pattern of the N_2O fluxes, they greatly lowered some peaks of N_2O emissions in the SX and HLJ vegetable soils (Fig. 2b and d).

Clearly, the NO fluxes demonstrated similar seasonal dynamics to the N_2O fluxes (Fig. 3). Some relatively high peak NO fluxes were still observed in the Spinach and Coriander herb planting seasons even though relatively low temperatures occurred during these periods, primarily due to lower soil moisture which was suitable for NO production.

The NO fluxes ranged from -44.6 to 377.6 μg N m^{-2} h^{-1} across all soil types. Furthermore, some NO peaks were significantly weakened with the Bw and Bm in the HN soil (Fig. 3a).

3.3. Cumulative N_2O, NO and NH_3 emissions

Cumulative N_2O emissions varied greatly among soil types (Table 2, $p < 0.001$), from 1.97 to 31.56 kg N ha^{-1} across all the soils during the vegetable cultivation period (Table 3a). Biochar amendments had significant influences on the cumulative N_2O emissions, reducing N_2O emissions by 13.7–41.6 % (Table 2). In comparison with the N treatment, biochar amendment resulted in no consistent effects on N_2O emissions over all soils (Table 3a), indicating significant interactions between biochar and soil types (Table 2, $p < 0.001$). Additionally, Bw amendment performed better mitigation effects which decreased N_2O emissions by 11.8–38.4 % across all the soils, significantly in HN soil in relation to Bm (Table 3a, $p < 0.05$). The values of cumulative NO emissions were much smaller than those of N_2O emissions, with a remarkable variation of 0.20–8.99 kg N ha^{-1} across all soils (Table 3b). Though pronounced effects on NO emissions with a reduction by average of 45.8 % (Table 2, $p < 0.05$), biochar amendments had no consensus effects across soils (Table 3b), which suggested significant interactions between biochar and soil types (Table 2, $p < 0.001$). Compared with Bm, Bw amendment significantly reduced NO emissions in SX and HLJ soils (Table 3b, $p < 0.05$). Moreover,, N_2O emissions had positive relationships with DEA both in SX and HLJ soils, and were affected positively with PNR in HN soil (Table 4). Additionally, NO emissions had positive correlations with both PNR and DEA in SX soil. However, neither N_2O nor NO emissions were influenced significantly by PNR and DEA in SD soils.

As is shown in Table 3c, the cumulative NH_3 emissions fluctuated greatly from 4.72–7.57 kg N ha^{-1}across all the soils. Though significantly enhancing NH_3 emissions (Table 2), biochar amendments produced no significant influences on the NH_3 emissions relative to N treatment in most soils (Table 3c). A tendency was found for the cumulative NH_3 emissions in N+Bm to be higher than those in the N+Bw treatment, although this difference was not remarkable within each soil. Additionally, stimulation effects were consistently present after the first fertilization event in each type of soil (Fig. 4).

3.4. Vegetable yield and gaseous reactive N intensity during the five-vegetable crop rotation

The vegetable yields for the five consecutive vegetable crops are presented in Table 3e. Pronounced differences existed among all soils (Table 2, $p < 0.001$). Biochar amendments exerted no significant effects on vegetable yield (Table 2). Compared with the N treatment, biochar amendments were prone to increase vegetable yield in SD and HLJ soils against HN and SX soils (Tables 3e), denoting pronounced interactions between soil and biochar (Table 2, $p < 0.05$). Compared with Bm, Bw amendment lowed total yield over all the soils (Table 3e), significantly in HN and SD soils ($p <$ 0.05).

Table 3f presents the GNrI during the whole experiment period, with a pronounced variation among soils (Table 2, $p < 0.001$). The GNrI was greatly affected by biochar amendment during the whole experiment period (Table 2, $p < 0.01$). Compared to N treatment, biochar amendments reduced the GNrI by 4.3–27.8 % across all soils, significantly in SX and HLJ soils (Table 3f, $p < 0.05$). Moreover, there were no remarkable differences between Bw and Bm throughout all soils.

4. Discussion

4.1. Biochar effects on GNrEs across different soil types

The effects of biochar amendment on the N_2O and NO emissions may be positive, negative or neutral, largely depending on the soil condition and the inherent characteristics of the biochar (Spokas and Reicosky, 2009; Nelissen et al., 2014). In our study, effects of two biochars on the N_2O and NO emissions did not follow a consensus trend across the four typical vegetable soils (Table 3a, b). In agreement with Cayuela et al. (2014), who reported that the role of biochar in mitigating N_2O emission was maximal in soils close to neutrality, remarkable mitigation effects were observed in SX and HLJ with the biochar amendments (Table 3a). These findings potentially resulted from the effects of the biochars on soil aeration, C/N ratio and pH, which affected the N dynamics and N cycling processes (Zhang et al., 2010; Ameloot et al., 2015). In line with Obia et al. (2015), biochar decreased NO emissions in low-pH HN soil (Table 3b), probably by inducing denitrification enzymes with higher activity, and then resulted in less NO accumulation relative to N_2 production. Moreover, the liming effects of biochar prevented the chemical decomposition of NO_2^- to NO (Islam et al., 2008), leaving only enzymatically produced NO to accumulate. However, neither N_2O nor NO emission was significantly influenced by PNR or DEA, suggesting other processes might play vital roles in SD soil. Besides nitrification and denitrification, nitrifier denitrification (Wrage et al., 2001) and heterotrophic nitrification (Zhu et al., 2011) can be important processes for producing N_2O and NO as well, especially in vegetable soils with low pH, low carbon content and high N content (Wrage et al., 2001). Ma et al. (2015) indicated that nitrifier denitrification might be the main process producing N_2O in the North China Plain. In addition, surplus N input in vegetable systems probably masked the beneficial effects of the biochar addition on the N transformation (Wang et al., 2015a). Therefore, the underlying mechanism of how biochar affect those processes needs to be illustrated in the further research.

On the other hand, different biochars may not produce universal influences on N_2O emissions for the same soil due to the distinct properties of the biochar (Spokas and Reicosky, 2009). In the current study, overall, in comparison with Bm, the Bw amendment had more effective mitigation effects on N_2O and NO emissions (Table 3a, b), largely due to the following reasons. First, compared with Bw, the contents of the TN and DOC in Bm were 1.8- and 1.4-fold (Table S1), respectively, which might supply extra N or C source for heterotrophic nitrification in the acidic HN soil, which made Bm ineffective for reducing the N_2O emissions (Table 3a). This result was in accordance with Li et al. (2015a), who observed that biochar amendment had no significant influence on the cumulative N_2O emissions, and even higher N_2O emissions occurred when biochar was input. Additionally, as shown in Fig.1, Bm was more prone to stimulate PNR and DEA, thus displaying lower mitigation ability than Bw. Second, compared with Bm, the C/N ratio was approximately twofold in Bw (Table S1), presumably leading to more inorganic nitrogen being immobilized in biochar with a higher

C/N ratio (Ameloot et al., 2015), decreasing the available N for microorganisms. Last, as presented in Fig. S3 and Table S1, Bw had more pores and surface area, having a better advantage over Bm in absorbing NO accordingly. Others have found that the lower mitigation capacity of high-N biochars (e.g., manures or biosolids) is probably due to the increased N release in the soil from the biochar (Schouten et al., 2012). To our knowledge, very few studies have investigated biochar effects on NO emissions (Nelissen et al., 2014; Obia et al., 2015), and the mechanisms through which biochar influence NO emissions are not elucidated yet. Therefore, more research is needed to clarify the underlying mechanisms of biochar on NO emission.

Intensive managed soils receiving fertilizer such as urea or anhydrous NH_3 and ruminant urine patches are potential hot spots for NH_3 formation, where the use of biochar is expected to retain NH_3–N in the soil system (Clough and Condron, 2010). Actually, the effects of biochar amendments on NH_3 volatilization largely depend on soil characteristics, biochar types and duration time. Soil texture is an important factor impacting NH_3 transfer and release. More clay contents were present in the SX soil (Table S1), which was limited in large soil pores, thus, the addition of porous biochar could enhance the soil aeration, promoting NH_3 volatilization (Sun et al., 2014). Additionally, it was worthy to note that cumulative NH_3 emissions were slightly higher in soils with the Bm than those with the Bw amendment (Fig. 4 and Table 3c) and that difference could presumably be attributed to less surface area and the much higher pH of Bm (Fig. S3 and Table S1), resulting in weak adsorption and great liming effects.

4.2. Biochar effects on vegetable yield and GNrI across different soil types

The application of biochar is usually intended to increase crop yields, and evidence suggests this may be successful (Schulz et al., 2013; Li et al., 2016). Due to its liming effect, biochar helps to improve the supply of essential macro- and micronutrients for plant growth (Chan and Xu, 2009; Major et al., 2010). Enhancement of vegetable yield with biochar amendment occurred in SD and HLJ soils (Table 3e). Additionally, the effects of Bm and Bw on vegetable yield were inconsistent, which probably due to the wide diversity of physicochemical characteristics of biochar that translates into variable reactions in soil (Novak et al., 2014). First, compared to Bw, Bm has a higher DOC content (Table S1), through which more nutrients may be directly introduced to the soil (Rajkovich et al., 2012). Secondly, besides their large amount of plant-available nutrients (Hass et al., 2012), biochars produced with manure have been generally considered significant for improving soil fertility by promoting soil structure development (Joseph et al., 2010), with the result that Bm was found superior to Bw in vegetable production enhancement in our case (Table 3e). As biochar effects on vegetable yield were variable, both biochar properties and soil conditions and crop species ought to be taken into account comprehensively before applying biochar to a certain soil condition.

However, no promotion of yield was observed with biochar amendments in HN and SX. This could be attributed to exacerbated soil salinity, which inhibited the uptake of nutrients and water (Ju et al., 2006; Zhou et al., 2010) and the growth of the soil microorganisms (Setia et al., 2011), leading to unsustainable greenhouse vegetable production. Compared with other biochar (Jia et al., 2012), the higher amounts of ash in Bw and Bm may contain high salts causing soil salinity (Hussain et al., 2016). After the addition of the two salt-rich biochars, the EC values of HN and SX vegetable soils increased and reached the limits to tolerance for the leafy vegetables (Shannon and Grieve, 1998). Here, we assessed two feedstock-derived biochar effects on GNrI in typical cultivated vegetable soils across mainland China. Overall, biochar amendments reduced GNrI over all the soils, with the magnitude largely depending on soil type. Remarkable reduction in GNrI had been detected due to the efficient mitigation induced by biochar in SX and HLJ (Table 3f). However, despite enhanced vegetable yield, no significant decreases in GNrI were observed in SD, mainly because of the absence of mitigation effects on N_2O, NO and NH_3 emissions of biochars (Table 3a, b and c) Overall, Bw was superior to Bm in mitigating the GNrE while Bm performed better in vegetable yield enhancement (Table 3d and e). Therefore, mitigation efficacys on GNrI were not notably different between Bw and Bm amendments across the four soils.

5. Conclusion

The study demonstrated that biochar amendments mostly reduced N_2O and NO emissions while slightly increased the NH_3 emissions, while produced no consensus influences on yield though those effects were largely both biochar- and soil-specific. Additionally, biochar amendments did decrease GNrI in intensive vegetable soils across mainland China. Furthermore, Bw was superior to Bm in mitigating the GNrE whereas the Bm performed better in crop yield throughout all soils. Consequently, both soil type and biochar characteristics need to be seriously considered before large-scale biochar application under certain regions of intensive vegetable production.

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References

Ameloot, N., Sleutel, S., Das, K. C., Kanagaratnam, J., and Neve, S. D.: Biochar amendment to soils with contrasting organic matter level: effects on N mineralization and biological soil properties, Global Change Biology Bioenergy, 7, 135–144, 2015.

Anderson, J. and Domsch, K.: A physiological method for the quantitative measurement of microbial biomass in soils, Soil biology and biochemistry, 10, 215–221, 1978.

Anenberg, S. C., Schwartz, J., Shindell, D., Amann, M., Faluvegi, G., Klimont, Z., Janssensmaenhout, G., Pozzoli, L., Van, D. R., and Vignati, E.: Global Air Quality and Health Co-benefits of Mitigating Near-Term Climate Change through Methane and Black Carbon Emission Controls, Environmental Health Perspectives, 120, 831–839, 2012.

Avnery, S., Mauzerall, D. L., Liu, J., and Horowitz, L. W.: Global crop yield reductions due to surface ozone exposure: 1. Year 2000 crop production losses and economic damage, Atmospheric Environment, 45, 2284–2296, 2011.

Behera, S. N., Sharma, M., Aneja, V. P., and Balasubramanian, R.: Ammonia in the atmosphere: a review on emission sources, atmospheric chemistry and deposition on terrestrial bodies, Environmental Science and Pollution Research, 20, 8092–8131, 2013.

Boyer, E. W., Goodale, C. L., Jaworski, N. A., and Howarth, R. W.: Anthropogenic nitrogen sources and relationships to riverine nitrogen export in the northeastern USA. In: The Nitrogen Cycle at Regional to Global Scales, Springer, 2002.

Cayuela, M., Van Zwieten, L., Singh, B., Jeffery, S., Roig, A., and Sánchez-Monedero, M.: Biochar's role in mitigating soil nitrous oxide emissions: A review and meta-analysis, Agriculture, Ecosystems & Environment, 191, 5–16, 2014.

Chan, K. Y. and Xu, Z.: Biochar: nutrient properties and their enhancement, Biochar for environmental management: science and technology, 2009. 67–84, 2009.

Ciais, P.: Carbon and other biogeochemical cycles: Final draft underlying scientific technical assessment, IPCC Secretariat, Geneva, 2013. 2013.

Clough, T. J. and Condron, L. M.: Biochar and the nitrogen cycle: introduction, Journal of Environmental Quality, 39, 1218-1223, 2010.

Clough, T. J., Bertram, J. E., Ray, J. L., Condron, L. M., O'Callaghan, M., Sherlock, R. R., and Wells, N. S.: Unweathered Wood Biochar Impact on Nitrous Oxide Emissions from a Bovine-Urine-Amended Pasture Soil, Soil Science Society of America Journal, 74, 852–860, 2010.

Deng, J., Zhou, Z., Zheng, X., and Li, C.: Modeling impacts of fertilization alternatives on nitrous oxide and nitric oxide emissions from conventional vegetable fields in southeastern China, Atmospheric Environment, 81, 642–650, 2013.

Diao, T., Xie, L., Guo, L., Yan, H., Lin, M., Zhang, H., Lin, J., and Lin, E.: Measurements of N_2O emissions from different vegetable fields on the North China Plain, Atmospheric Environment, 72, 70–76, 2013.

FAO, IIASA, ISRIC, and ISSCAS: Harmonized World Soil Database Version 1.2, 2012. 2012.

Field, J. L., Keske, C. M. H., Birch, G. L., Defoort, M. W., and Cotrufo, M. F.: Distributed biochar and bioenergy coproduction: a regionally specific case study of environmental benefits and economic impacts, Global Change Biology Bioenergy, 5, 177–191, 2013.

Food and Agriculture Organization (FAO) (2015) FAOSTAT (Food and Agriculture Organization Statistical Data) Statistical Yearbook Vol. 4. Available at: http: // faostat.fao.org (accessed 12 August 2015)

Harrison, R. and Webb, J.: A review of the effect of N fertilizer type on gaseous emissions, Advances in Agronomy, 73, 65–108, 2001.

Hass, A., Gonzalez, J. M., Lima, I. M., Godwin, H. W., Halvorson, J. J., and Boyer, D. G.: Chicken manure biochar as liming and nutrient source for acid Appalachian soil, Journal of Environmental Quality, 41, 1096–1106, 2012.

Hu, H. W., Macdonald, C. A., Trivedi, P., Anderson, I. C., Zheng, Y., Holmes, B., Bodrossy, L., Wang, J. T., He, J. Z., and Singh, B. K.: Effects of climate warming and elevated CO_2 on autotrophic nitrification and nitrifiers in dryland ecosystems, Soil Biology & Biochemistry, 92, 1–15, 2016.

Hussain, M., Farooq, M., Nawaz, A., Al-Sadi, A. M., Solaiman, Z. M., Alghamdi, S. S., Ammara, U., Yong, S. O., and Siddique, K. H. M.: Biochar for crop production: potential benefits and risks, Journal of Soils & Sediments, 2016. 1–32, 2016.

IPCC: Climate Change 2013: The Physical Science Basis: working group I contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Stockholm, 2013.

Islam, A., Chen, D., White, R. E., and Weatherley, A. J.: Chemical decomposition and fixation of nitrite in acidic pasture soils and implications for measurement of nitrification, Soil Biology & Biochemistry, 40, 262–265, 2008.

Jia, J., Li, B., Chen, Z., Xie, Z., and Xiong, Z.: Effects of biochar application on vegetable production and emissions of N_2O and CH_4, Soil Science and Plant Nutrition, 58, 503–509, 2012.

Joseph, S. D., Campsarbestain, M., Lin, Y., Munroe, P., Chia, C. H., Hook, J., Van, Z. L., Kimber, S., Cowie, A., and Singh, B. P.: An investigation into the reactions of biochar in soil, Australian Journal of Soil Research, 48, 501–515, 2010.

Ju, X. T., Kou, C. L., Zhang, F. S., and Christie, P.: Nitrogen balance and groundwater nitrate contamination: comparison among three intensive cropping systems on the North China Plain, Environmental Pollution, 143, 117–125, 2006.

Kim, J. Y., Song, C. H., Ghim, Y. S., Won, J. G., Yoon, S. C., Carmichael, G. R., and Woo, J. H.: An investigation on NH_3

emissions and particulate NH_4^+–NO_3^- formation in East Asia, Atmospheric Environment, 40, 2139–2150, 2006.

Kurola, J., Salkinoja-Salonen, M., Aarnio, T., Hultman, J., and Romantschuk, M.: Activity, diversity and population size of ammonia-oxidising bacteria in oil-contaminated landfarming soil, FEMS Microbiology Letters, 250, 33–38, 2005.

Langridge, J. M., Lack, D., Brock, C. A., Bahreini, R., Middlebrook, A. M., Neuman, J. A., Nowak, J. B., Perring, A. E., Schwarz, J. P., and Spackman, J. R.: Evolution of aerosol properties impacting visibility and direct climate forcing in an ammonia-rich urban environment, Journal of Geophysical Research Atmospheres, 117, 2240–2260, 2012.

Li, B., Bi, Z., and Xiong, Z.: Dynamic responses of nitrous oxide emission and nitrogen use efficiency to nitrogen and biochar amendment in an intensified vegetable field in southeastern China, Global Change Biology Bioenergy, 2016. 2016.

Li, B., Fan, C. H., Xiong, Z. Q., Li, Q. L., and Zhang, M.: The combined effects of nitrification inhibitor and biochar incorporation on yield-scaled N_2O emissions from an intensively managed vegetable field in southeastern China, Biogeosciences, 12, 15185–15214, 2015a.

Li, B., Fan, C. H., Zhang, H., Chen, Z. Z., Sun, L. Y., and Xiong, Z. Q.: Combined effects of nitrogen fertilization and biochar on the net global warming potential, greenhouse gas intensity and net ecosystem economic budget in intensive vegetable agriculture in southeastern China, Atmospheric Environment, 100, 10–19, 2015b.

Liu, X., Zhang, A., Ji, C., Joseph, S., Bian, R., Li, L., Pan, G., and Paz-Ferreiro, J.: Biochar's effect on crop productivity and the dependence on experimental conditions—a meta-analysis of literature data, Plant and Soil, 373, 583–594, 2013.

Lu, R.: Methods of soil and agro-chemical analysis, China Agricultural Science and Technology Press, Beijing, 2000. 127–332, 2000. (in Chinese)

Ma, L., Shan, J., Yan, X., 2015. Nitrite behavior accounts for the nitrous oxide peaks following fertilization in a fluvo-aquic soil. Biology and Fertility of Soils 51, 563-572.

Major, J., Lehmann, J., Rondon, M., and Goodale, C.: Fate of soil-applied black carbon: downward migration, leaching and soil respiration, Global Change Biology, 16, 1366–1379, 2010.

Mei, B. L., Zheng, X. H., Xie, B. H., Dong, H. B., Zhou, Z. X., Rui, W., Jia, D., Feng, C., Tong, H. J., and Zhu, J. G.: Nitric oxide emissions from conventional vegetable fields in southeastern China, Atmospheric Environment, 43, 2762–2769, 2009.

Misselbrook, T. H., Weerden, T. J. V. D., Pain, B. F., Jarvis, S. C., Chambers, B. J., Smith, K. A., Phillips, V. R., and Demmers, T. G. M.: Ammonia emission factors for UK agriculture, Atmospheric Environment, 34, 871–880(810), 2000.

Mukherjee, A. and Zimmerman, A. R.: Organic carbon and nutrient release from a range of laboratory-produced biochars and biochar–soil mixtures, Geoderma, s 193–194, 122–130, 2013.

Nelissen, V.: Effect of different biochar and fertilizer types on N_2O and NO emissions, Soil Biology & Biochemistry, 70, 244–255, 2014.

Novak, J. M., Spokas, K. A., Cantrell, K. B., Ro, K. S., Watts, D. W., Glaz, B., Busscher, W. J., and Hunt, P. G.: Effects of biochars and hydrochars produced from lignocellulosic and animal manure on fertility of a Mollisol and Entisol, Soil Use and Management, 30, 175–181, 2014.

Obia, A., Cornelissen, G., Mulder, J., and Dörsch, P.: Effect of Soil pH Increase by Biochar on NO, N_2O and N_2 Production during Denitrification in Acid Soils, Plos One, 10, 359–367, 2015.

Pacholski, A., Cai, G. X., Fan, X. H., Ding, H., Chen, D., Nieder, R., and Roelcke, M.: Comparison of different methods for the measurement of ammonia volatilization after urea application in Henan Province, China, Journal of Plant Nutrition and Soil Science, 171, 361–369, 2008.

Pinder, R. W., Adams, P. J., and Pandis, S. N.: Ammonia emission controls as a cost-effective strategy for reducing atmospheric particulate matter in the Eastern United States, Environmental Science & Technology, 41, 380–386, 2007.

Powlson, D. S., Addiscott, T. M., Benjamin, N., Cassman, K. G., de Kok, T. M., Van, G. H., L'Hirondel, J. L., Avery, A. A., and Van, K. C.: When does nitrate become a risk for humans?, Journal of Environmental Quality, 37, 291–295, 2008.

Rajkovich, S., Enders, A., Hanley, K., Hyland, C., Zimmerman, A. R., and Lehmann, J.: Corn growth and nitrogen nutrition after additions of biochars with varying properties to a temperate soil, Biology & Fertility of Soils, 48, 271–284, 2012.

Ravishankara, A. R., Daniel, J. S., and Portmann, R. W.: Nitrous oxide (N_2O): the dominant ozone-depleting substance emitted in the 21st century, Science, 326, 123–125, 2009.

Saarnio, S., Heimonen, K., and Kettunen, R.: Biochar addition indirectly affects N_2O emissions via soil moisture and plant N uptake, Soil Biology & Biochemistry, 58, 99–106, 2013.

Schouten, S., Groenigen, J. W. V., Oenema, O., and Cayuela, M. L.: Bioenergy from cattle manure? Implications of anaerobic digestion and subsequent pyrolysis for carbon and nitrogen dynamics in soil, Global Change Biology Bioenergy, 4, 751–760, 2012.

Schulz, H., Dunst, G., and Glaser, B.: Positive effects of composted biochar on plant growth and soil fertility, Agronomy for Sustainable Development, 33, 817–827, 2013.

Setia, R., Marschner, P., Baldock, J., Chittleborough, D., and Verma, V.: Relationships between carbon dioxide emission and soil properties in salt-affected landscapes, Soil Biology & Biochemistry, 43, 667–674, 2011.

Shannon, M. C. and Grieve, C. M.: Tolerance of vegetable crops to salinity, Scientia Horticulturae, 78, 5–38, 1998.

Smith, M. S. and Tiedje, J. M.: Phases of denitrification following oxygen depletion in soil, Soil Biology & Biochemistry, 11, 261–267, 1979.

Smith, P.: Soil carbon sequestration and biochar as negative emission technologies, Global Change Biology, 51, 574–575, 2016.

Sohi, S. P.: Agriculture. Carbon storage with benefits, Science, 338, 1034–1035, 2012.

Sororzano, L.: Determination of ammonia in natural waters by the phenolhypochlorite method, Limnol. Oceanogr, 14, 799–801, 1969.

Spokas, K. A. and Reicosky, D. C.: Impacts of sixteen different biochars on soil greenhouse gas production, Ann. Environ. Sci, 3, 4, 2009.

Stavi, I. and Lal, R.: Agroforestry and biochar to offset climate change: a review, Agronomy for Sustainable Development, 33, 81–96, 2013.

Sun, L., Li, L., Chen, Z., Wang, J., and Xiong, Z.: Combined effects of nitrogen deposition and biochar application on emissions of N_2O, CO_2 and NH_3 from agricultural and forest soils, Soil science and plant nutrition, 60, 254–265, 2014.

Ussiri, D. and Lal, R.: The Role of Nitrous Oxide on Climate Change, Springer Netherlands, 2013.

Wang, J., Chen, Z., Ma, Y., Sun, L., Xiong, Z., Huang, Q., and Sheng, Q.: Methane and nitrous oxide emissions as affected by organic–inorganic mixed fertilizer from a rice paddy in southeast China, Journal of Soils and Sediments, 13, 1408–1417, 2013.

Wang, J., Chen, Z., Xiong, Z., Chen, C., Xu, X., Zhou, Q., and Kuzyakov, Y.: Effects of biochar amendment on greenhouse gas emissions, net ecosystem carbon budget and properties of an acidic soil under intensive vegetable production, Soil Use and Management, 31, 375–383, 2015a.

Wang, S., Nan, J., Shi, C., Fu, Q., Gao, S., Wang, D., Cui, H., Saizlopez, A., and Zhou, B.: Atmospheric ammonia and its impacts on regional air quality over the megacity of Shanghai, China, Scientific Reports, 5, 2015b.

Wrage, N., Velthof, G., Van Beusichem, M., Oenema, O., 2001. Role of nitrifier denitrification in the production of nitrous oxide. Soil Biology and Biochemistry 33, 1723-1732.

Xiong, Z., Xie, Y., Xing, G., Zhu, Z., and Butenhoff, C.: Measurements of nitrous oxide emissions from vegetable production in China, Atmospheric Environment, 40, 2225–2234, 2006.

Yao, Z., Zheng, X., Xie, B., Mei, B., Wang, R., Klaus, B. B., Zhu, J., and Yin, R.: Tillage and crop residue management significantly affects N-trace gas emissions during the non-rice season of a subtropical rice-wheat rotation, Soil Biology & Biochemistry, 41, 2131–2140, 2009.

Zhang, A., Cui, L., Pan, G., Li, L., Hussain, Q., Zhang, X., Zheng, J., and Crowley, D.: Effect of biochar amendment on yield and methane and nitrous oxide emissions from a rice paddy from Tai Lake plain, China, Agriculture Ecosystems & Environment, 139, 469–475, 2010.

Zhang, F., Chen, X., and Vitousek, P.: Chinese agriculture: An experiment for the world, Nature, 497, 33-35, 2013.

Zhang, M., Fan, C. H., Li, Q. L., Li, B., Zhu, Y. Y., and Xiong, Z. Q.: A 2-yr field assessment of the effects of chemical and biological nitrification inhibitors on nitrous oxide emissions and nitrogen use efficiency in an intensively managed vegetable cropping system, Agriculture Ecosystems & Environment, 201, 43–50, 2015.

Zhang, Y., Luan, S., Chen, L., and Shao, M.: Estimating the volatilization of ammonia from synthetic nitrogenous fertilizers used in China, Journal of Environmental Management, 92, 480–493, 2011.

Zhao, L. M., Wu, L. H., Dong, C. J., and Li, Y. S.: Rice yield, nitrogen utilization and ammonia volatilization as influenced by modified rice cultivation at varying nitrogen rates, Agricultural Sciences, 01, 10–16, 2010.

Zhao, X., Wang, J., Wang, S., and Xing, G.: Successive straw biochar application as a strategy to sequester carbon and improve fertility: A pot experiment with two rice/wheat rotations in paddy soil, Plant and Soil, 378, 279–294, 2014.

Zheng, X., Mei, B., Wang, Y., Xie, B., Wang, Y., Dong, H., Xu, H., Chen, G., Cai, Z., and Yue, J.: Quantification of N_2O fluxes from soil–plant systems may be biased by the applied gas chromatograph methodology, Plant and Soil, 311, 211–234, 2008.

Zhou, J. B., Chen, Z. J., Liu, X. J., Zhai, B. N., and Powlson, D. S.: Nitrate accumulation in soil profiles under seasonally open ‘sunlight greenhouses’ in northwest China and potential for leaching loss during summer fallow, Soil Use and Management, 26, 332–339, 2010.

Zhu, T., Zhang, J., Cai, Z., 2011. The contribution of nitrogen transformation processes to total N_2O emissions from soils used for intensive vegetable cultivation. Plant and Soil 343, 313-327.

Table legends

Table 1

Soil organic carbon (SOC), soil total nitrogen (TN), soil pH, electric conductivity (EC) and microbial biomass carbon (MBC) as affected by different treatments across the four vegetable soils.

Soil	Treatment	SOC (g kg^{-1})	TN (g kg^{-1})	pH	EC (ds m^{-1})	MBC (mg kg^{-1})
HN	N	8.0 ± 0.8c	1.37 ± 0.12b	4.37 ± 0.04c	1.76 ± 0.21b	1353 ± 119a
	N+Bw	15.6 ± 0.5b	1.47 ± 0.07b	4.64 ± 0.04b	2.43 ± 0.31a	1173 ± 49b
	N+Bm	18.8 ± 0.6a	1.64 ± 0.04a	5.01 ± 0.03a	2.00 ± 0.32ab	1234 ± 50ab
SX	N	9.7 ± 0.7c	1.55 ± 0.04b	7.53 ± 0.02b	1.74 ± 0.27b	490 ± 9a
	N+Bw	15.6 ± 0.8b	1.62 ± 0.06b	7.61 ± 0.05a	2.25 ± 0.22a	495 ± 16a
	N+Bm	17.5 ± 1.1a	1.79 ± 0.03a	7.63 ± 0.01a	1.96 ± 0.06ab	504 ± 18a
SD	N	7.9 ± 0.1b	1.13 ± 0.04b	7.70 ± 0.08a	0.85 ± 0.03b	535 ± 13b
	N+Bw	14.2 ± 0.6a	1.20 ± 0.04b	7.66 ± 0.03a	0.92 ± 0.04a	554 ± 10ab
	N+Bm	15.5 ± 1.4a	1.37 ± 0.06a	7.71 ± 0.03a	0.87 ± 0.02ab	573 ± 12a
HLJ	N	29.9 ± 0.5b	2.19 ± 0.04b	6.91 ± 0.05a	0.83 ± 0.03b	921 ± 44b
	N+Bw	36.0 ± 1.5a	2.20 ± 0.03b	6.92 ± 0.06a	0.95 ± 0.03a	988 ± 56b
	N+Bm	38.1 ± 1.8a	2.41 ± 0.01a	6.94 ± 0.04a	0.92 ± 0.06a	1242 ± 196a
ANOVA results						
Biochar		***	***	***	***	*
Soil		***	***	***	***	***
Biochar × Soil		*	n.s.	***	n.s.	**

Data shown are means ± standard deviations of three replicates. See Fig. 1 for treatments codes. Different letters within the same column indicate significant differences among treatments within the same soil at $p < 0.05$ level.

***Significant at $p < 0.001$; **significant at $p < 0.01$; *significant at $p < 0.05$; n.s. not significant.

Table 2

Two-way ANOVA and mean effects of biochar (Bc) and soil (S) types on cumulative N_2O, NO and NH_3 emissions, gaseous reactive nitrogen emission (GNrE), vegetable yield and gaseous reactive nitrogen intensity (GNrI) during the entire sampling period.

Factors	DF	N_2O emission			NO emission			NH_3 emission			GNrE			Vegetable yield			GNrI		
		SS	F	P	SS	F	P	SS	F	P	SS	F	P	SS	F	P	SS	F	P
Bc	2	271.9	65.1	***	46.4	174.7	***	0.5	0.8	n.s.	380.5	86.4	***	76.2	3.2	n.s.	0.1	7.9	**
S	3	1429.9	228.1	***	152.2	382.1	***	4.1	3.8	*	2322.6	351.5	***	4316.9	123.3	***	2.3	110.3	***
Bc×S	6	179.3	14.3	***	33.4	41.9	***	1.4	0.7	n.s.	234.5	17.7	***	230.4	3.3	*	0.1	1.6	n.s.
Model	11	4009.7	174.5	***	225.3	154.3	***	29.1	7.5	***	5290	218.3	***	15962.0	124.4	***	5.8	77.0	***
Error	24	50.1			3.2			8.5			52.9			280.0			0.2		
biochar effect (n = 9)																			
N mean			12.01 ±1.44a			2.86 ±0.24a			5.92 ±0.24b			43.81 ±1.25b			20.50 ±1.60a			0.57 ±0.05a	
N+Bw mean			7.01 ±0.58b			1.55 ±0.14b			6.65 ±0.27a			43.53 ±1.67b			14.94 ±0.84b			0.45 ±0.04b	
N+Bm mean			10.37 ±0.56a			1.55 ±0.10b			7.01 ±0.25a			49.53 ±1.11a			18.60 ±0.65a			0.49 ±0.03ab	
Soil effect (n = 9)																			
HN mean			27.20 ±1.85a			5.80 ±0.50a			5.31 ±0.16c			33.06 ±1.65c			38.04 ±1.90a			1.15 ±0.11a	
SX mean			4.89 ±0.45b			1.08 ±0.13b			12.69 ±0.46a			25.05 ±1.11d			12.69 ±0.46b			0.51 ±0.01b	
SD mean			2.25 ±0.26c			0.25 ±0.09c			9.51 ±0.55b			44.88 ±0.49b			9.51 ±0.55c			0.21 ±0.01c	
HLJ mean			4.48 ±0.68b			0.81 ±0.04b			11.79 ±0.71a			79.50 ±2.41a			11.79 ±0.71b			0.15 ±0.01c	

SS: the sum of squares.

F value: the ratio of mean squares of two independents samples.

P value: the index of differences between the control group and the experimental group. *, ** and *** indicate significance at $p < 0.05$, $p < 0.01$ and $p < 0.001$, respectively.

n.s.: not significant.

Data shown are means ± standard deviations of the nine replicates. See Fig. 1 for treatments codes. Different letters within the same column indicate significant differences among treatments at $p < 0.05$ level.

Table 3

Cumulative gaseous nitrogen (N_2O, NO and NH_3) emissions, gaseous reactive nitrogen emission (GNrE), vegetable yield and gaseous reactive nitrogen intensity (GNrI) under the different treatments across the four soils.

Treatments	HN	SX	SD	HLJ
(a) Cumulative N_2O emissions (kg N ha^{-1})				
N	30.59±3.15a	7.83±0.60a	2.52±0.37a	7.10±1.91a
N+Bw	19.45±2.43b	3.20±0.28b	1.97±0.21a	3.45±0.86b
N+Bm	31.56±1.35a	3.63±0.62b	2.26±0.58a	4.01±0.68b
(b) Cumulative NO emissions (kg N ha^{-1})				
N	8.99±1.01a	1.27±0.15a	0.20±0.08a	0.97±0.11a
N+Bw	4.54±0.60b	0.80±0.13b	0.33±0.19a	0.52±0.03b
N+Bm	3.87±0.30b	1.16±0.17a	0.21±0.10a	0.94±0.03a
(c) Cumulative NH_3 emissions (kg N ha^{-1})				
N	4.72±0.27a	5.79±0.54b	6.34±0.51a	5.67±0.42a
N+Bw	5.09±0.38a	6.83±0.74ab	7.35±0.75a	6.24±0.49a
N+Bm	5.32±0.42a	7.57±0.57a	7.37±1.11a	6.48±0.43a
(d) GNrE (kg N ha^{-1})				
N	44.30±3.13a	14.89±1.33a	9.06±0.80a	13.74±1.67a
N+Bw	29.08±2.21b	10.82±1.14b	9.64±0.88a	10.21±0.92b
N+Bm	40.76±1.66a	12.36±0.74b	9.84±0.49a	11.42±0.27b
(e) Vegetable yield (t ha^{-1})				
N	35.20±2.52a	25.29±3.90a	39.09±2.03b	75.65±5.84b
N+Bw	29.05±2.35b	23.57±1.74a	44.53±3.74b	76.95±4.04ab
N+Bm	34.93±2.87a	26.30±2.63a	51.00±3.18a	85.89±3.29a
(f) GNrI (kg N t^{-1} yield)				
N	1.27±0.18a	0.59±0.08a	0.23±0.02a	0.18±0.04a
N+Bw	1.01±0.12a	0.46±0.05b	0.22±0.04a	0.13±0.02b
N+Bm	1.17±0.15a	0.47±0.04b	0.19±0.01a	0.13±0.01b

Data shown are means ± standard deviations of the three replicates. See Fig. 1 for treatments codes. Different letters within the same column indicate significant differences among treatments within the same soil at $p < 0.05$ level.

Table 4

The correlations between N_2O or NO emission and PNR or DEA in each soil.

Item	HN		SX		SD		HLJ	
	PNR	DEA	PNR	DEA	PNR	DEA	PNR	DEA
N_2O	0.75*	0.66	0.49	0.76*	-0.10	0.16	-0.82**	0.70*
NO	0.62	-0.29	0.79*	0.69*	-0.54	0.01	-0.63	0.22

Asterisks indicated 0.05 level significances (*$p < 0.05$) and 0.01 level significances (**$p < 0.01$), n = 9.

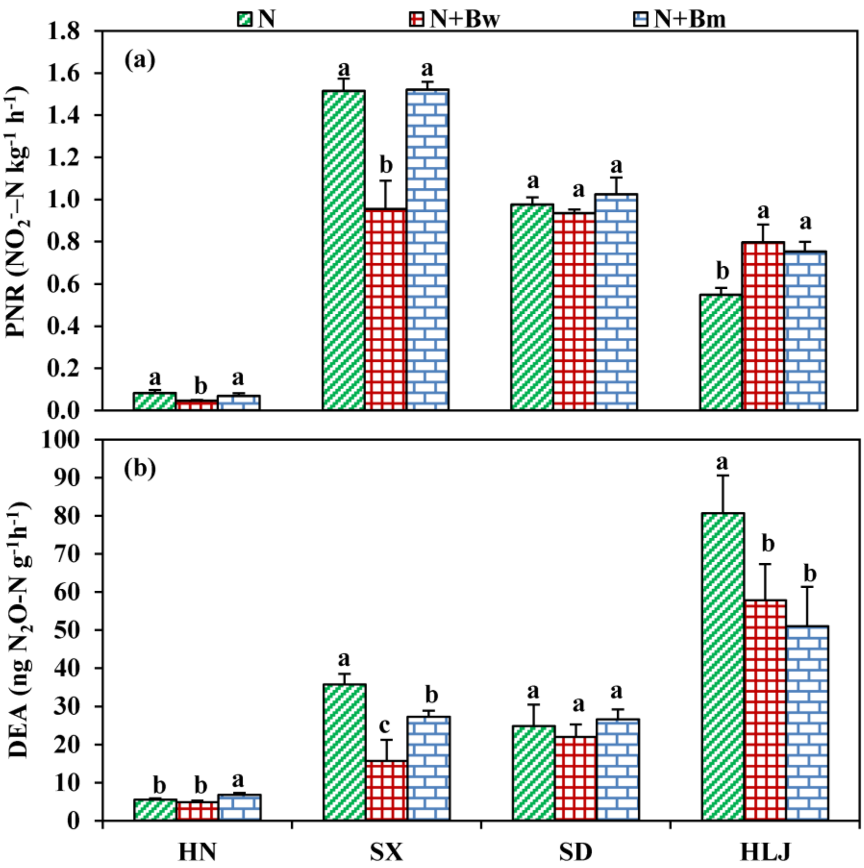
Figure legends

Fig. 1 Potential nitrification rate (PNR) and Denitrification enzyme activity (DEA) under different treatments in HN, SX, SD and HLJ soils. The three treatments with each soil were urea without biochar (N), urea with wheat straw biochar (N+Bw) and urea with swine manure biochar (N+Bm). Bars indicate standard deviation (mean + SD, n = 3). Different letters above the bars indicate significant differences among the different treatments within the same soil, at $p < 0.05$.

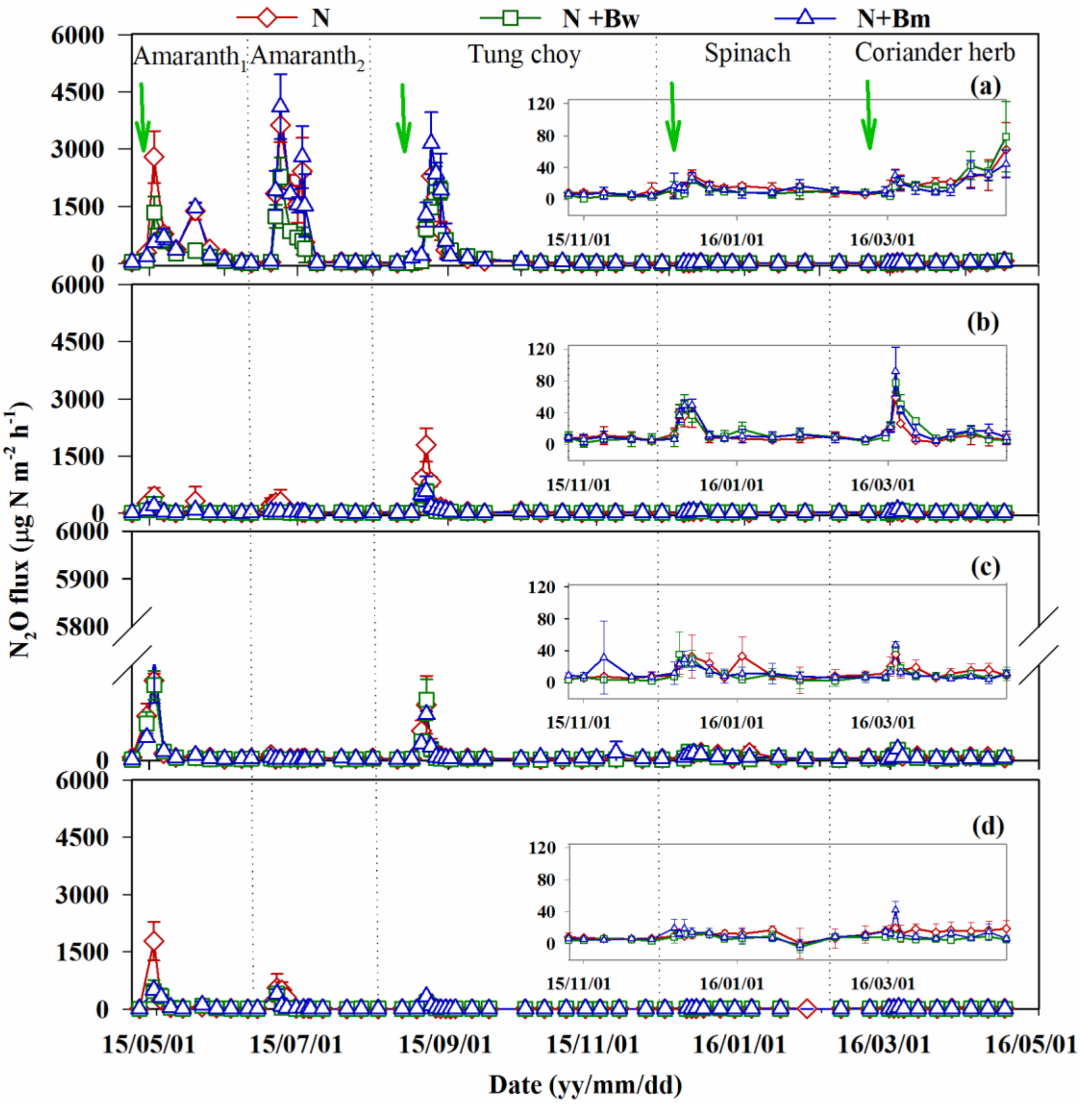
Fig. 2 Temporal dynamics of soil N_2O (μg N m^{-2} h^{-1} ± SD, n = 3) fluxes under different treatments in HN (a), SX (b), SD (c) and HLJ (d) vegetable soils with five consecutive vegetable crops. The solid arrows indicate fertilization. See Fig. 1 for treatments codes.

Fig. 3 Temporal dynamics of soil NO (μg N m^{-2} h^{-1} ± SD, n = 3) fluxes under different treatments in HN (a), SX (b), SD (c) and HLJ (d) vegetable soils with five consecutive vegetable crops. The solid arrows indicate fertilization. See Fig. 1 for treatments codes.

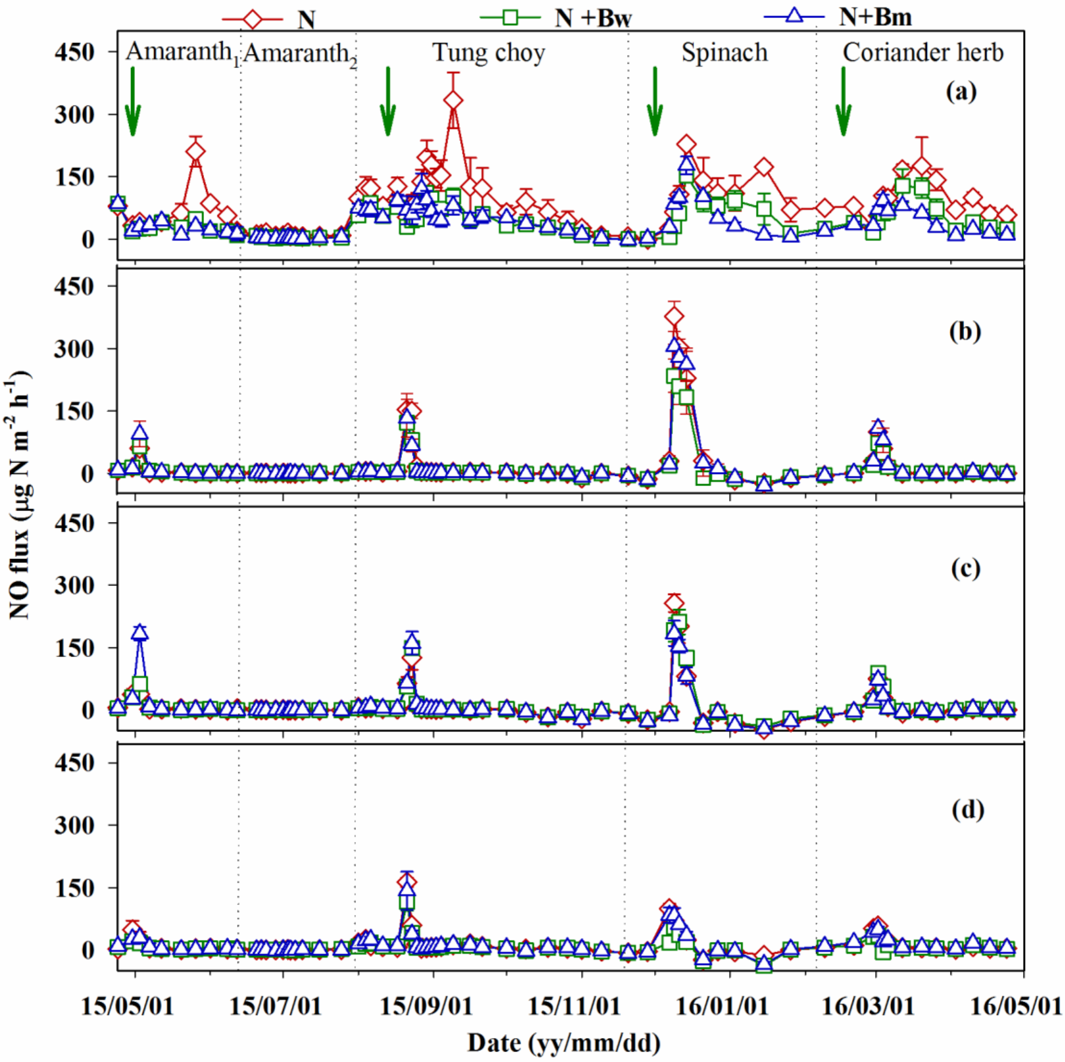
Fig. 4 Cumulative ammonia (NH_3) emissions from the HN (a), SX (b), SD (c) and HLJ (d) soils during the four nitrogen fertilization events F: every N fertilization event. The bars indicate the standard deviation of the mean (kg N ha^{-1} ± SD, n = 3) of each treatment for the sum of the four N fertilization events. See Fig. 1 for treatments codes. Different letters above the bars indicate significant differences among the different treatments for each soil, at $p < 0.05$.


N
N+Bw
N+Bm
(a)
PNR (NO2−-N kg-1 h-1)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
(b)
DEA (ng N2O-N g-1h-1)
0
10
20
30
40
50
60
70
80
90
100
HN
SX
SD
HLJ

1

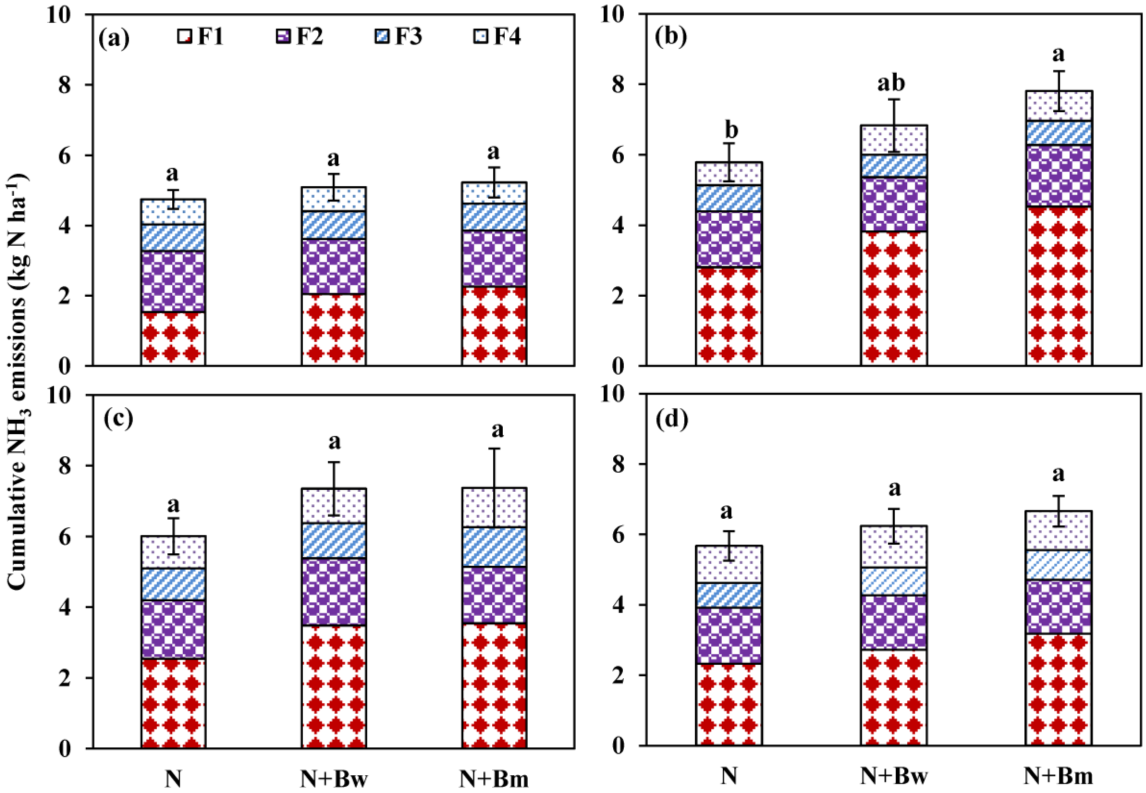

N
N +Bw
N+Bm
Amaranth1
Amaranth2
Tung choy
Spinach
Coriander herb
(a)
(b)
(c)
(d)
N2O flux (μg N m-2 h-1)
Date (yy/mm/dd)
15/05/01
15/07/01
15/09/01
15/11/01
16/01/01
16/03/01
16/05/01

1


N
N +Bw
N+Bm
Amaranth$_1$
Amaranth$_2$
Tung choy
Spinach
Coriander herb
(a)
(b)
(c)
(d)
450
300
150
0
NO flux (μg N m^{-2} h^{-1})
15/05/01
15/07/01
15/09/01
15/11/01
16/01/01
16/03/01
16/05/01
Date (yy/mm/dd)

1

1



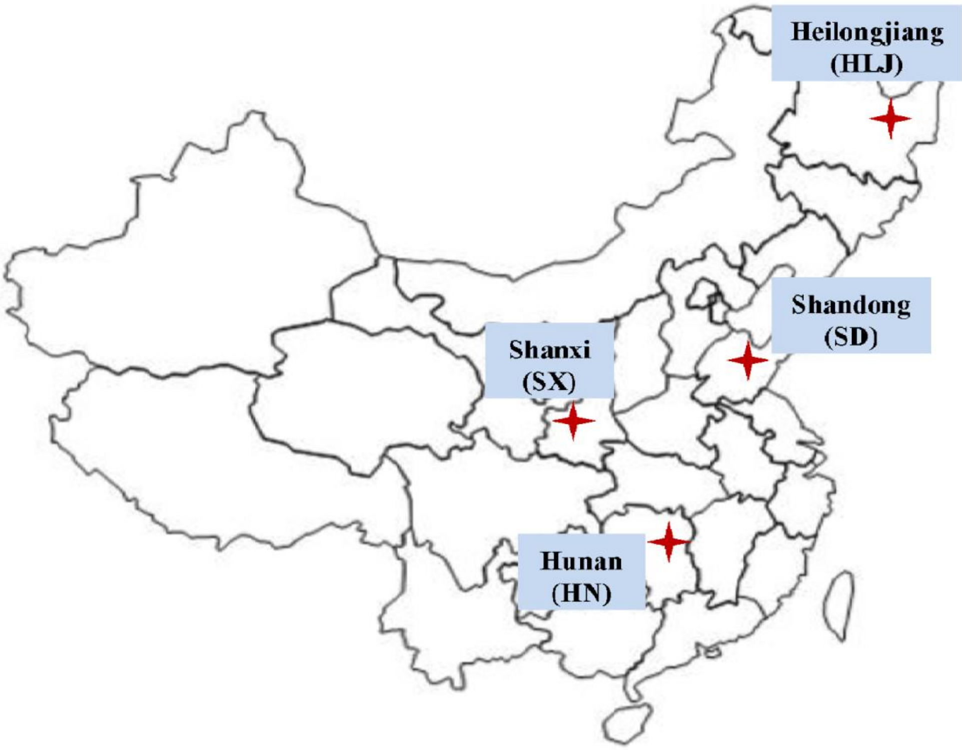
(a)
F1
F2
F3
F4
(b)
(c)
(d)
Cumulative NH3 emissions (kg N ha-1)
N
N+Bw
N+Bm
a
b
ab

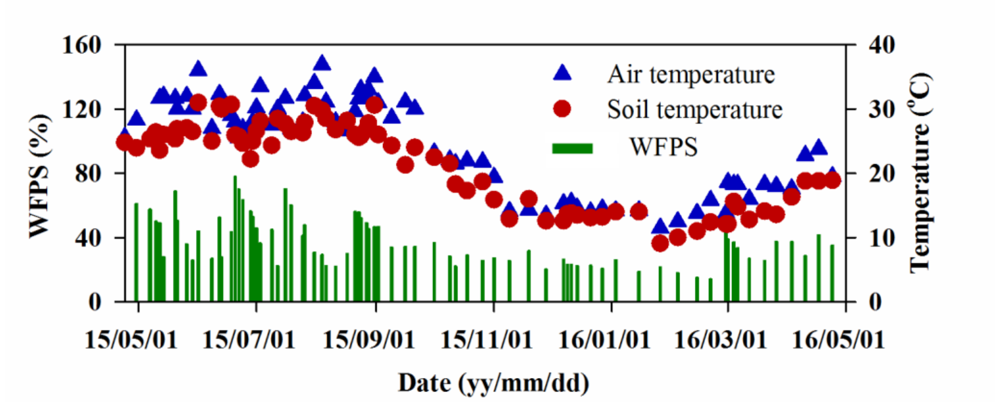
Supplementary information

Fig. S1 Map showing the sampling sites in China.

Fig. S2 Dynamics of water filled pore space (WFPS), air temperature and soil temperature during the vegetable cultivation period.

Fig. S3 Scanning electron microscope (SEM) images of the biochars derived from Bw (a, b and c) and Bm (d, e and f). Same magnification for a and d (×50), b and e (×400) and c and f (×2000).





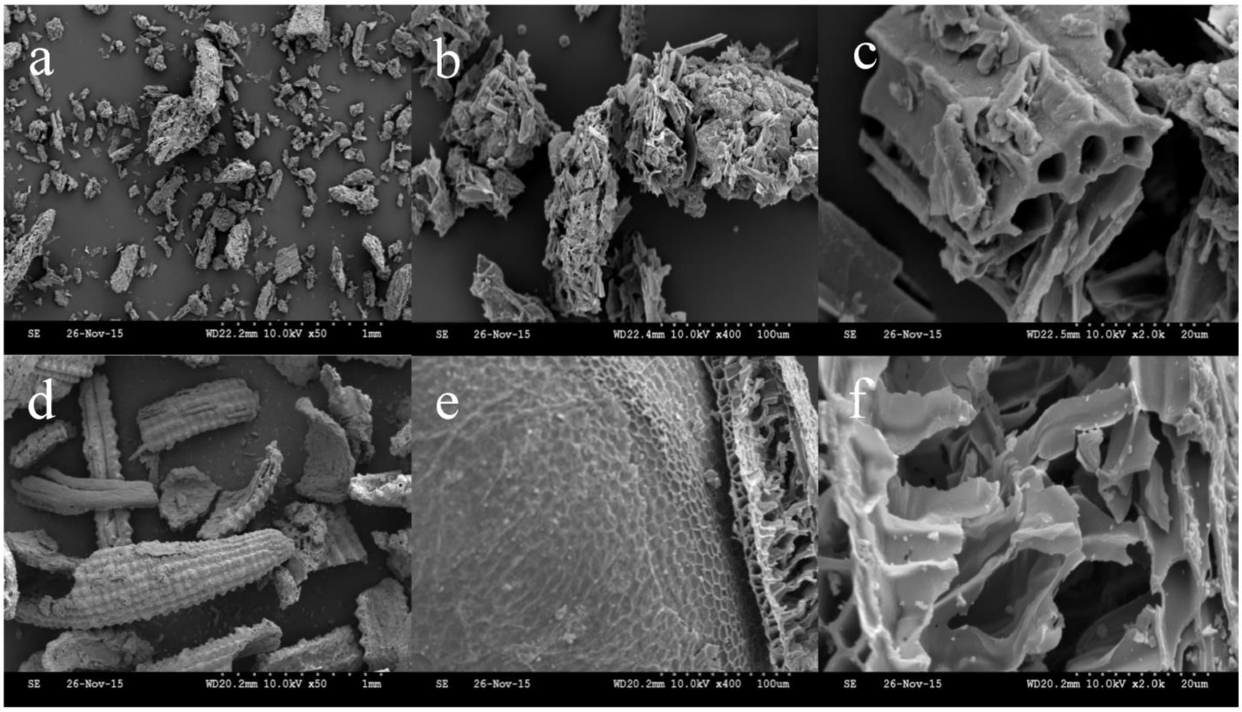

a
SE 26-Nov-15 WD22.2mm 10.0kV x50 1mm
b
SE 26-Nov-15 WD22.4mm 10.0kV x400 100um
c
SE 26-Nov-15 WD22.5mm 10.0kV x2.0k 20um
d
SE 26-Nov-15 WD20.2mm 10.0kV x50 1mm
e
SE 26-Nov-15 WD20.2mm 10.0kV x400 100um
f
SE 26-Nov-15 WD20.2mm 10.0kV x2.0k 20um

Table S1

Characteristics of the vegetable soils and biochars used in the experiment.

Iterm	Vegetable soil				Biochar	
	HN	SX	SD	HLJ	Bw	Bm
Texture	sandy loam	silt (sandy) clay loam	silt (sandy) loam	silt (sandy) loam		
sand, %	47.1	17.7	24.7	31.6		
silt, %	40.0	59.6	60.4	52.8		
clay, %	12.9	22.7	14.9	15.6		
total C (g kg^{-1})	7.6	9.8	8.2	26.8	449.1	461.2.
total N (g kg^{-1})	1.2	1.4	1.0	2.1	6.5	12.0
C/N	6.3	7.0	8.2	12.8	69.1	38.4
H (g kg^{-1})					10.5	16.1
O (g kg^{-1})					52.4	96.7
H/Corg					0.3	0.4
pH	5.6	7.6	8.2	7.6	9.7	10.0
EC (ds m^{-1})	1.8	1.1	0.2	0.2	10.6	3.3
DOC (g kg^{-1})	0.5	0.4	0.2	0.7	0.9	1.3
CEC, cmol kg^{-1}	6.1	13.2	15.3	20.3	22.1	22.7
WHC, %	41.6	50.1	54.4	59.6	362.0	304.1
NH_4^+–N (mg kg^{-1})	105.3	32.2	28.4	31.6	4.3	4.0
NO_3^-–N (mg kg^{-1})	415.8	307.6	21.2	30.8	6.1	3.2
Bulk density (g cm^{-3})	1.2	1.4	1.1	1.1		
Surface area (m^2 g^{-1})					21.3	9.3
Ash content, %					29.1	38.6

EC: electronic conductivity; DOC: dissolved organic carbon; CEC: cation exchange capacity; WHC: water holding capacity

Table S2

Crop rotation, tillage practices, and fertilizer application from April 2015 to April 2016.

Crop	Date	Agricultural activity	Fertilizer N rate (kg N ha^{-1})	Fertilizer P rate (kg N ha^{-1})	Fertilizer K rate (kg N ha^{-1})
$Amaranth_1$	04/22/2015	Tillage			
	04/29/2015	Fertilizer application and planting	240	240	240
	06/13/2015	Harvesting			
	06/14/2015	Tillage			
$Amaranth_2$	06/19/2015	Fertilizer application and planting	0	0	0
	07/31/2015	Harvesting			
	07/32/2015	Tillage			
Tung choy	08/20/2015	Fertilizer application and planting	200	200	200
	11/27/2015	Harvesting			
	11/28/2015	Tillage			
Spinach	12/06/2015	Fertilizer application and planting	150	150	150
	01/28/2016	Harvesting			
	01/09/2016	Tillage			
Coriander herb	02/28/2016	Fertilizer application and planting	180	180	180
	04/29/2016	Harvesting			
	04/30/2016	Tillage			

Biochar can decrease the gaseous reactive nitrogen intensity in intensive vegetable soils across mainland China~~Effects of two contrasting biochars on gaseous nitrogen emissions and intensity in intensive vegetable soils across mainland China~~

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Tel: +86-25-84395148; Fax: +86-25-84395210

Highlights

1. Two contrasting biochars affected G~~aseous ~~Nr~~itrogen ~~I~~ntensity~~ across 4 major vegetable soils in China.
2. Biochar affects gaseous~~-~~ Nr or yield largely depending on soil types.
3. Both biochars decreased GNrI with Bw mitigated gaseous Nr whereas Bm improved yield.

Abstract

Biochar amendment to soil has been proposed as a strategy for sequestering carbon, mitigating climate change and enhancing crop productivity, but few studies have demonstrated the general effects of different feedstock-derived biochars on the various gaseous reactive nitrogen emissions (GNrEs, N_2O, NO and NH_3) simultaneously across the typical vegetable soils in China. A greenhouse pot experiment with five consecutive vegetable crops was conducted to investigate the effects of two contrasting biochar, namely, wheat straw biochar (Bw) and swine manure biochar (Bm) on GNrEs, vegetable yield and gaseous reactive nitrogen intensity (GNrI) in four typical vegetable soils from ~~the main vegetable production regions (~~Hunan province (HN), Shanxi province (SX), Shandong province (SD) and Heilongjiang province (HLJ) which~~) that~~ are representative of the intensive vegetable ecosystems across mainland China. Results showed that remarkable GNrE mitigation induced by biochar occurred in SX and HLJ soils, whereas enhancement of yield occurred in SD and HLJ soils. Additionally, both biochars decreased GNrI with Bw performed better than Bm regarding N_2O mitigation, with Bw mitigating~~ed~~ N_2O and NO emissions by 21.8–59.1 % and 37.0–49.5 % (except for SD), respectively, while Bm improved yield by ~~4.0~~13.5–30.5 % (except for HN and SX).~~—~~Biochar amendments generally stimulated the NH_3 emissions with greater enhancement from –Bm than Bw. We can infer that ~~Since~~ the biochar's effects on the GNrEs and vegetable yield strongly depend~~ed~~ on the attributes of the soil and biochar.~~,~~ Therefore, both soil type and biochar characteristics should be seriously considered before conducting large-scale application of biochar in order to achieve the maximum benefits under intensive greenhouse vegetable agriculture.

Keyword: Biochar, Intensive vegetable soil, Gaseous reactive nitrogen emissions (GNrEs), Gaseous reactive nitrogen intensity (GNrI)

带格式的：下标

1 Introduction

Agriculture accounted for an estimated emission of 4.1 (1.7–4.8) Tg N yr^{-1} for N_2O and 3.7 Tg N yr^{-1} for NO, contributing 60 % and 10 %, respectively, to the total global anthropogenic emissions, largely due to increases of N fertilizer application in cropland (Ciais, 2013). The concentration of atmospheric N_2O, a powerful, long-lived, greenhouse gas, has increased from 270 parts per billion by volume (ppbv) in the pre-industrial era to ~ 324 ppbv (Ussiri and Lal, 2013); it has ~~298~~ 265 times the global warming potential (GWP) of CO_2 on a 100-year horizon (IPCC, 2013) and also causes depletion of the ozone layer in the atmosphere (Ravishankara et al., 2009). In contrast, NO_x, which is mainly emitted as nitric oxide (NO), does not directly affect the earth's radiative balance but catalyzes the production of tropospheric ozone (O_3), which is a greenhouse gas associated with detrimental effects on human health (Anenberg et al., 2012) and crop production (Avnery et al., 2011). Additionally, along with the high nitrogen (N) application, ammonia volatilization is one of the major N loss pathways (Harrison and Webb, 2001) as well, with up to 90% coming from agricultural activities (Misselbrook et al., 2000; Boyer et al., 2002). As a natural component and a dominant atmospheric alkaline gas, NH_3 plays an important role in atmospheric chemistry and ambient aerosol formation (Langridge et al., 2012; Wang et al., 2015b). In addition to nutrient enrichment (eutrophication) of terrestrial and aquatic systems and global acidification of precipitation, NH_3 has also been shown to be a major factor in the formation of atmospheric particulate matter and secondary aerosols (Kim et al., 2006; Pinder et al., 2007), leading to potentially adverse effects on human and ecosystem health such as visibility degradation and threats to biodiversity (Powlson et al., 2008; Behera et al., 2013). Consequently, the release of various reactive N species results in lower N use efficiency in agricultural systems.

In China, vegetable production devotes an area of approximately 24.7 $\times 10^6$ ha, equivalent to 12.4% of the total available cropping area, and the production represented 52 % of the world vegetable production in 2012 (FAO, 2015). Intensified vegetable cultivation in China is characterized by high N application rates, high cropping index and frequent farm practices. Annual nitrogen fertilizer inputs for intensively managed vegetable cultivation in rapidly developing areas are 3–6 times higher than in cereal grain cultivation in China (Ju et al., 2006; Diao et al., 2013; Wang et al., 2015a). As a result, great concern exists about excess N fertilizer application, leading to low use efficiency in intensive vegetable fields in China (Deng et al., 2013; Diao et al., 2013). Meanwhile, intensive vegetable agriculture is considered to be an important source of N_2O (Xiong et al., 2006; Jia et al., 2012; Li et al., 2015b; Zhang et al., 2015) and NO production (Mei et al., 2009). Moreover, ammonia volatilization is another important N pathway in fertilized soil, resulting in large losses of soil-plant N (Pacholski et al., 2008; Zhang et al., 2011). Therefore, the reduction of reactive N loss becomes a central environmental challenge to meet the joint challenges of high production and acceptable environmental consequences in intensive vegetable production (Zhang et al., 2013).

Biochar is the dark-colored, carbon (C)-rich residue of pyrolysis or gasification of plant biomass under oxygen (O_2)-limited conditions, specifically produced for use as a soil amendment (Sohi, 2012). The amendment of agricultural ecosystems with biochar has been proposed as an effective countermeasure for climate change (Smith, 2016). These additions would increase soil carbon storage (Mukherjee and Zimmerman, 2013; Stavi and Lal, 2013), decrease GHG emissions (Li et al., 2016), and improve soil fertility and crop production (Major et al., 2010; Liu et al., 2013). However, some recent studies have reported no difference or even an increase in soil N_2O emissions induced by biochar application from different soils (Saarnio et al., 2013; Wang et al., 2015a). Still, NH_3 volatilization was enhanced by biochar application in pasture soil (Clough et al., 2010), vegetable soil (Sun et al., 2014) and paddy soil in the wheat-growing season (Zhao et al., 2014). Additionally, crop productivity responses to biochar amendments differed among various biochars (Cayuela et al., 2014). These inconsistent results suggest that current biochar application to soil is not a "one-size fit-all paradigm" because of the variation in the physical and chemical characteristics of the different biochars, soil types and crop species (Field et al., 2013; Cayuela et al., 2014). Moreover, limited types of biochar (Spokas and Reicosky, 2009) and soil (Sun et al., 2014) were involved in the experiments in previous studies. Thus, the evaluation of the different types of biochar under the typical soils is imperative to gain a comprehensive understanding of potential interactions before the large-scale application of biochars in intensive vegetable cropping system in China.

Therefore, a greenhouse pot experiment was conducted in an effort to investigate the effects of different types of biochar on gaseous reactive nitrogen emissions (GNrEs), namely, N_2O, NO and NH_3, simultaneously in four typical intensified vegetable soils across main vegetable production areas of mainland China. Overall, the objectives of this research were to gain a comprehensive insight into the effects of the different types of biochar on the GNrEs, vegetable yield and gaseous reactive nitrogen intensity (GNrI) in intensively managed vegetable production in China.

2 Materials and methods

2.1. Experimental soil and biochar

Four typical greenhouse vegetable cultivation sites with a long history (more than 10 years) of conventional cultivation were selected from Northeast, Northwest, Central and Eastern China (Fig. S1), namely, Phaeozem, Anthrosol, Acrisol and Cambisol (FAO and ISRIC, 2012) from Jiamusi (46°48´N, 130°12´E), Heilongjiang province (HLJ); Yangling (34°18´N, 108°2´E), Shanxi province (SX); Changsha (28°32´N, 113°23´E), Hunan province (HN) and Shouguang (36°56´N, 118°38´E), Shandong province (SD), respectively were collected and represented a range of differences in physicochemical properties and regions (Table S1). Soil samples were manually collected from the cultivated layer (0–20 cm) after the local vegetable harvest in April, 2015. The samples were air-dried and passed through a 5 mm stainless steel mesh sieve and homogenized thoroughly. Any visible roots and organic residues were removed manually before being packed with the necessary amount of soil to achieve the initial field bulk density. Each pot received 15 kg of 105 ℃ dry-weight-equivalent fresh soil. For each of the biochar amendment pot~~s~~, 282.6 g pot^{-1} sieved biochar (2 mm) was mixed with the soil thoroughly before the experiment, which was equivalent to a 40 t ha^{-1} biochar dose (dry weight). No more biochar was added later in the experimental period.

Two types of biochar, derived from two common agricultural wastes in China: wheat straw and swine manure, hereafter referred to as Bw and Bm, respectively (Table S1). The Bw was produced at the Sanli New Energy Company in Henan, China, by pyrolysis and thermal decomposition at 400–500 ℃. The Bm was produced through thermal decomposition at 400 ℃ by the State Key Laboratory of Soil Science and Sustainable Agricultural, Institute of Soil Science, Chinese Academy of Sciences. In accordance with Lu (2000), the SOC was measured by wet digestion with H_2SO_4–$K_2Cr_2O_7$, TN was determined by semi-micro Kjeldahl digestion, and soil texture was determined with the pipette method. The soil pH and biochar pH were measured in deionized water at a volume ratio of 1:2.5 (soil to water) with a PHS-3C mv/pH detector (Shanghai Kangyi Inc. China). Biochar content of hydrogen (H) was measured by elemental analysis after dry combustion (Euro EA, Hekatech GmbH, Wegberg, Germany). The oxygen content of biochar was measured with the same device after pyrolysis of the sample at 1000 ℃ followed by reduction of the evolved O_2 to CO and quantification by GC-TCD. The soil NO_3^-–N and NH_4^+–N were measured following the two-wavelength ultraviolet spectrometry and indophenol blue methods, respectively, using an ultraviolet spectrophotometer (HITACHI, UV-2900, Tokyo, Japan). Electric conductivity (EC) was measured by using a Mettler-Toledo instrument (FE30-K, Shanghai, China) at a 1:5 (w:v) soil to water ratio. Cation exchange capacity (CEC) was determined using the CH_3COONH_4 method. Dissolved organic carbon (DOC) was extracted from 5 g of the biochar/soil with an addition of 50 ml deionized water and measured by a TOC analyzer (TOC-2000/3000, Metash Instruments Co., LTD, Shanghai, China). Ash content was measured by heating the biochars at 750 ℃ for 4 h. The specific surface area of the biochar material was tested using the Brunauer–Emmett–Teller (BET) method, from which the N adsorption–desorption isotherms at 77 K were measured by an automated gas adsorption analyzer ASAP2000 (Micromeritics, Norcross, GA) with + 5% accuracy. Scanning electron microscopy (SEM) imaging analysis was conducted using a HITACHI S-3000N scanning electron microscope.

2.2. Experimental set-up and management

The pot experiments were performed at the greenhouse experimental station of Nanjing Agricultural University, China. Five vegetable crops were grown successively in the four vegetable soils during the experimental period. For each type of soil, three treatments with three replicates were arranged in a ~~completely~~ random design: urea without biochar (N), urea with wheat straw biochar (N+Bw), urea with swine manure biochar (N+Bm). In addition, phosphate and potassium fertilizers in the form of calcium magnesium phosphate and potassium chloride, together with urea, were broadcasted and mixed with soil thoroughly prior to sowing the vegetables. No topdressing events occurred because of the frequent cultivation and short growth period for the leafy vegetables. Based on the vegetable growth, all pots received equal amounts of water and no precipitation. Detailed information on the pot management practices is provided in Table S2.

Each pot consists of a 30 cm × 30 cm (height × diameter) cylinder made of polyvinyl chloride (PVC). The top of each pot was surrounded by a special water-filled trough collar, which allowed a chamber to sit on the pot and prevent gas exchange during the gas-sampling period. Small holes (diameter of 1 cm) at the bottom of the pots were designed for drainage. To prevent soil loss, a fine nylon mesh (< 0.5 mm) was attached to the base of the soil cores before packing.

2.3. Measurement of N_2O, NO and NH_3

The NO and N_2O fluxes were measured simultaneously from each vegetable cultivation using a static opaque chamber method (Zheng et al., 2008; Yao et al., 2009). A square PVC chamber of 35 cm × 35 cm × 40 cm (length × width × height) was temporarily mounted on the pot for gas flux measurement. The chamber was coated with sponge and aluminum foil outside to prevent solar radiation heating the chamber. Gas samples for flux measurements were collected between 8 and 10 a.m. on each measuring day to minimize the influence of diurnal temperature variation. Gas fluxes were usually measured once a week and every other day for one week following fertilizer application. To measure the N_2O flux, four samples were collected from the headspace chamber using 20 ml polypropylene syringes at 0, 10, 20, and 30 min after chamber closure. The gas concentrations in the samples were analyzed within 12 h after sampling using an Agilent 7890A gas chromatograph equipped with an electron capture detector (ECD) for N_2O detection. The carrier gas was argon-methane (50 %) at a flow rate of 40 ml min^{-1}. The column and ECD temperatures were maintained at 40 and 300 ℃, respectively. The gas chromatography configurations described by Wang et al. (2013) were adopted for the gas concentration analysis. N_2O flux was calculated using the linear increases in gas concentration with time. Sample sets were rejected unless they yielded a linear regression value of $R^2 > 0.90$.

For each NO flux measurement, gas samples were collected from the same chamber that was used for the N_2O flux measurements (Yao et al., 2009). Before closing the chamber, an approximately 1.0 L gas sample from the headspace of each chamber was extracted into an evacuated sampling bag (Delin Gas Packing Co., LTD, Dalian, China), and this measurement was regarded as time 0 min for NO analysis. After 30 min under chamber enclosure conditions (i.e., after the N_2O sample collections were completed), another headspace gas sample with the same volume was extracted from each chamber into another evacuated bag. Within 1 h after sampling, NO concentrations were analyzed by a model 42*i* chemiluminescence NO–NO–NO_X analyzer (Thermo Environmental Instruments Inc., Franklin, MA, USA). The NO fluxes were derived from the concentration differences between the two collected samples. The NOx analyzer was calibrated by a model 146*i* dynamic dilution calibrator system at the end of each crop-growing season.

The mean flux of N_2O or NO during the experiment period was calculated as the average of all measured fluxes, which were weighted by the interval between the two measurements (Xiong et al., 2006). The cumulative N_2O was calculated as the product of the mean flux and the entire duration.

The NH_3 volatilization was determined using the ventilation method (Zhao et al., 2010). The phosphoglycerol-soaked sponge was replaced every day after each fertilization event for approximately one week. The phosphoglycerol-soaked sponges used to collect the NH_3 samples were immediately extracted with 300 mL potassium chloride (KCl) solution (1 mol L^{-1}) for 1 h. The concentration of ammonia nitrogen (NH_4^+–N) was measured using the indophenol blue method at 625 nm (Sororzano, 1969) by ultraviolet spectrophotometry (HITACHI, UV-2900, Tokyo, Japan, with 0.005 absorbance of photometric accuracy). The cumulative seasonal NH_3 volatilization was the sum of the daily emissions during the measurement period.

2.4. Auxiliary measurements

Simultaneously with the determination of trace gas fluxes, the air temperature and the soil temperature at a depth of 5 cm were measured using thermally sensitive probes at each sampling date. Soil water content was also measured using a portable water detector (Mode TZS-1K, Zhejiang Top Instrument Corporation Ltd., China) by the frequency domain reflectometer method at a depth of 5 cm. Measured soil water contents (v/v) were converted to water filled pore space (WFPS) with the following equation:

$$\text{WFPS} = \text{volumetric water content (cm}^3\text{ cm}^{-3}) / \text{total soil porosity (cm}^3\text{ cm}^{-3}) \quad (1)$$

Here, total soil porosity = [1 − (soil bulk density (g cm^{-3}) / 2.65)] with an assumed soil particle density of 2.65 (g cm^{-3}). The total soil bulk density was determined with the cutting ring method according to Lu (2000).

After each vegetable crop reached physiological maturity, the fresh vegetable yield was measured by weighing the whole aboveground and belowground biomass in each pot.

$$GNrE = \text{cumulative } N_2O + \text{cumulative NO} + \text{cumulative } NH_3 \text{ emissions (kg N ha}^{-1}) \quad (2)$$

$$GNrI = GNrE \,/\, \text{vegetable fresh yield (kg N t}^{-1} \text{ yield)} \quad (3)$$

After the one-year pot experiment, a soil sample from each pot was blended carefully. One subsample was stored at 4 ℃ for determination of microbial biomass carbon (MBC), potential nitrification rate (PNR) and denitrification enzyme activity (DEA) within 3 days. Another subsample was air-dried for analysis of SOC, TN, pH and EC. MBC was determined by substrate-induced respiration using a gas chromatography (Anderson and Domsch 1978). PNR was measured using the chlorate inhibition soil-slurry method as previously described (Kurola et al., 2005) with modifications (Hu et al., 2016). DEA was quantified as described by Smith and Tiedje (1979).

2.5. Data processing and statistics

One-way ANOVA was performed to test the effects of the treatments on cumulative N_2O, NO and NH_3 emissions; GNrE; vegetable yield and GNrI. Two-way ANOVA was used to analyze the effects of the biochar type; soil type; and their interactions on N_2O, NO and NH_3 emissions, vegetable yield, GNrE and GNrI throughout the experimental period. Multiple comparisons among the treatments were further explained using Tukey's HSD test. Significant differences were considered at $P < 0.05$. All statistical analyses were performed using JMP ver. 7.0 (SAS Institute, Cary, NC, USA, 2007). Pearson's correlation analysis was used to determine whether there were significant interrelationships between N_2O/NO and PNR or DEA in each soil, using SPSS window version 18.0 (SPSS Inc., Chicago, USA).

3. Results

3.1. Soil responses to biochar amendment

Obvious differences in all observed soil properties existed among soil types (Table 1, $p < 0.001$), suggesting the wide variations of soil characters across mainland China. Additionally, biochar amendments had significant influences on all the soil properties (Table 1, $p < 0.05$). Compared with N treatments, biochar amendments increased the SOC, TN and EC by 20.4–135.0 %, 0.5–21.2 % and 2.4–38.1 %, respectively, across all the soils. Compared with Bw, Bm amendment resulted in higher contents of SOC and TN by 5.8–20.5 % and 9.5–14.2 %, respectively, whereas EC values were higher by 3.3–21.5 % induced by Bw than Bm amendment over all soils. Additionally, biochar amendments significantly increased~~enhanced~~ soil pH by 0.27–0.64 and 0.08–0.10 units compared with N treatment in HN and SX soils ($p < 0.05$), respectively, and higher values were detected with Bm than Bw amendment in all soils. Furthermore, biochar amendments tended to increase MBC in SD and HLJ soils, and Bm performed better in MBC enhancements than Bw in all soils.

As shown in Fig. 1, no consensus effects on PNR and DEA were observed with biochar amendments across all soils. Compared with N treatment, biochar amendments significantly increased PNR in HLJ while exerted no influences on SD soil (Fig. 1a). Compared with Bw, Bm amendment significantly increased PNR in HN and SX soils. Moreover, compared with N, biochar amendments ~~significantly~~ reduced DEA in most soils, significantly in SX and HLJ by an average of 40.1 and 37.8 % ~~in SX and HLJ~~ (Fig. 1b, $p < 0.05$), respectively~~, while producing no influence in SD soils (Fig. 1b)~~. In comparison with Bw, remarkable enhancements in DEA were observed by 42.5 and 74.4 % with Bm amendment in HN and SX soils, respectively ($p < 0.05$).

3.2. Seasonal variations of N_2O and NO emissions

The dynamics of N_2O fluxes from all N-applied treatments in the four vegetable soils were relatively consistent and followed a sporadic and pulse-like pattern that was accompanied with fertilization, tillage and irrigation (Fig. 2). In addition, peak N_2O fluxes varied greatly. Most of the N_2O emissions occurred during the Amaranth and Tung choy growing periods, and there were several small emissions peaks during the Spinach and Coriander herb growing periods due to lower N application rate (Table S2), soil temperature and water content (Fig. S2). The highest peaks of N_2O emissions from HN, SX, SD and HLJ were 4133.7, 1784.0, 432.4 and 1777.2 μg N m^{-2} h^{-1}, respectively. Although biochar (Bw and Bm) application did not significantly alter the seasonal pattern of the N_2O fluxes, they greatly lowered some peaks of N_2O emissions in the SX and HLJ vegetable soils (Fig. 2b and d).

Clearly, the NO fluxes demonstrated similar seasonal dynamics to the N_2O fluxes (Fig. 3). Some relatively high peak NO fluxes were still observed in the Spinach and Coriander herb planting seasons even though relatively low temperatures occurred during these periods, primarily due to lower soil moisture which was suitable for NO production. The NO fluxes ranged from -44.6 to 377.6 μg N m^{-2} h^{-1} across all soil types. Furthermore, some NO peaks were significantly weakened with the Bw and Bm in the HN soil (Fig. 3a).

3.3. Cumulative N_2O, NO and NH_3 emissions

Cumulative N_2O emissions varied greatly among soil types (Table 2, $p < 0.001$), from 1.97 to 31.56 kg N ha^{-1} across all the soils during the vegetable cultivation period (Table 3a). Biochar amendments had significant influences on the cumulative N_2O emissions, reducing N_2O emissions by 13.7–41.6 % (Table 2). In comparison with the N treatment, biochar amendment resulted in no consistent effects on N_2O emissions over all soils ~~decreased N_2O emissions by an average of 56.4 % and 47.5 % in SX and HLJ~~ (Table 3a~~, $p < 0.05$~~), ~~respectively, with no remarkable influence in SD soil,~~ indicating significant interactions between biochar and soil types (Table 2, $p < 0.001$). Additionally,~~Compared with Bm,~~ Bw amendment performed better mitigation effects which decreased N_2O emissions by 11.8–38.4 % across all the soils, significantly in HN soil in relation to Bm (Table 3a, $p < 0.05$). ~~In comparison with N_2O emission, t~~The values of cumulative NO emissions ~~was~~ were much smaller than those of N_2O emissions, with a remarkable variation of 0.20–8.99 kg N ha^{-1} across all soils (Table 3b). Though pronounced effects on NO emissions with a reduction by average of 45.8 % (Table 2, $p < 0.05$~~-~~), biochar amendments had no consensus effects across soils~~, reducing NO emissions in HN soil~~ (Table 3b~~, $p < 0.05$~~)~~ and producing no remarkable influence on SD soil~~, which suggested significant interactions between biochar and soil types (Table 2, $p < 0.001$). Compared with Bm, Bw amendment significantly reduced NO emissions in SX and HLJ soils (Table 3b, $p < 0.05$). Moreover,~~As shown in Table 4~~, N_2O emissions had positive relationships with DEA both in SX and HLJ soils, and were affected positively with~~by~~ PNR in HN soil (Table 4). Additionally, NO emissions had positive correlations with both PNR and DEA in SX soil. However, neither N_2O nor NO emissions were influenced significantly by PNR and DEA in SD soils.

As is shown in Table 3c, the cumulative NH_3 emissions fluctuated greatly from 4.72–7.57 kg N ha^{-1}across all the soils. Though significantly enhancing NH_3 emissions (Table 2), biochar amendments produced no significant influences on the NH_3 emissions relative to N treatment in most soils (Table 3c). A tendency was found for the cumulative NH_3 emissions in N+Bm to be higher than those in the N+Bw treatment, although this difference was not remarkable within each soil. Additionally, stimulation effects were consistently present after the first fertilization event in each type of soil (Fig. 4).

3.4. Vegetable yield and gaseous reactive N ~~emissions~~ intensity during the five-vegetable crop rotation

The vegetable yields for the five consecutive vegetable crops are presented in Table 3e. Pronounced differences existed among all soils (Table 2, $p < 0.001$). Biochar amendments exerted no significant effects on vegetable yield (Table

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2). Compared with the N treatment, biochar amendments were prone to increase vegetable yield in SD and HLJ soils against HN and SX soils (Tables 3e), denoting pronounced interactions between soil and biochar (Table 2, $p < 0.05$). Compared with Bm, Bw amendment lowed total yield over all the soils (Table 3e), significantly in HN and SD soils ($p < 0.05$).

Table 3f presents the GNrI during the whole experiment period, with a pronounced variation among soils (Table 2, $p < 0.001$). The GNrI was greatly affected by biochar amendment during the whole experiment period (Table 2, $p < 0.01$). Compared to N treatment, biochar amendments reduced the GNrI by 4.3–27.8 % across all soils, significantly in SX and HLJ soils (Table 3f, $p < 0.05$). Moreover, there were no remarkable differences between Bw and Bm throughout all soils.

4. Discussion

4.1. Biochar effects on GNrEs across different soil types

The effects of biochar amendment on the N_2O and NO emissions may be positive, negative or neutral, largely depending on the soil condition and the inherent characteristics of the biochar (Spokas and Reicosky, 2009; Nelissen et al., 2014). In our study, effects of two biochars on the N_2O and NO emissions did not follow a consensus trend across the four typical vegetable soils (Table 3a, b). In agreement with Cayuela et al. (2014), who reported that the role of biochar in mitigating N_2O emission was maximal in soils close to neutrality, remarkable mitigation effects were observed in SX and HLJ with the biochar amendments (Table 3a). These findings potentially resulted from the effects of the biochars on soil aeration, C/N ratio and pH, which affected the N dynamics and N cycling processes (Zhang et al., 2010; Ameloot et al., 2015). ~~Moreover, mitigation of N_2O emissions induced by biochar was probably due to the decreased denitrification in SX and HLJ soils (Fig.1b and Table 4).~~ In line with Obia et al. (2015), biochar decreased NO emissions in low-pH HN soil (Table 3b), probably by inducing denitrification enzymes with higher activity, and then resulted in less NO accumulation relative to N_2 production. Moreover, the liming effects of biochar prevented the chemical decomposition of NO_2^- to NO (Islam et al., 2008), leaving only enzymatically produced NO to accumulate. However, neither N_2O nor NO emission was significantly influenced by PNR or DEA, suggesting other processes might play vital roles in SD soil. Besides nitrification and denitrification, nitrifier~~s~~ denitrification (Wrage et al., 2001) and heterotrophic nitrification (Zhu et al., 2011) can be important processes for producing N_2O~~/~~ and NO as well, especially in vegetable soils with low pH, low carbon content and high N content (Wrage et al., 2001). Ma et al. (2015) indicated that nitrifier denitrification might be the main process producing N_2O in the North China Plain. In addition, surplus N input in vegetable systems probably masked the beneficial effects of the biochar addition on the N transformation (Wang et al., 2015a). Therefore, the underlying mechanism of how biochar affect those processes needs to be illustrated in the further research.

On the other hand, different biochars may not produce universal influences on N_2O emissions for the same soil due to the distinct properties of the biochar (Spokas and Reicosky, 2009). In the current study, overall, in comparison with Bm, the Bw amendment had more effective mitigation effects on N_2O and NO emissions (Table 3a, b), largely due to the following reasons. First, compared with Bw, the contents of the TN and DOC in Bm were 1.8- and 1.4-fold (Table S1), respectively, which might supply extra N or C source for heterotrophic nitrification in the acidic HN soil, which made Bm ineffective for reducing the N_2O emissions (Table 3a). This result was in accordance with Li et al. (2015a), who observed that biochar amendment had no significant influence on the cumulative N_2O emissions, and even higher N_2O emissions occurred when biochar was input. Additionally, as shown in Fig.1, Bm was more prone to stimulate PNR and

DEA, thus displaying lower mitigation ability than Bw. Second, compared with Bm, the C/N ratio was approximately twofold in Bw (Table S1), presumably leading to more inorganic nitrogen being immobilized in biochar with a higher C/N ratio (Ameloot et al., 2015), decreasing the available N for microorganisms. Last, as presented in Fig. S3 and Table S1, Bw had more pores and surface area, having a better advantage over Bm in absorbing NO accordingly. Others have found that the lower mitigation capacity of high-N biochars (e.g., manures or biosolids) is probably due to the increased N release in the soil from the biochar (Schouten et al., 2012). To our knowledge, very few studies have investigated biochar effects on NO emissions (Nelissen et al., 2014; Obia et al., 2015), and the mechanisms through which biochar influence NO emissions are not elucidated yet. Therefore, more research is needed to clarify the underlying mechanisms of biochar on NO emission.

Intensive managed soils receiving fertilizer such as urea or anhydrous NH_3 and ruminant urine patches are potential hot spots for NH_3 formation, where the use of biochar is expected to retain NH_3–N in the soil system (Clough and Condron, 2010). Actually, the effects of biochar amendments on NH_3 volatilization largely depend on soil characteristics, biochar types and duration time. Soil texture is an important factor impacting NH_3 transfer and release. More clay contents were present in the SX soil (Table S1), which was limited in large soil pores, thus, the addition of porous biochar could enhance the soil aeration, promoting NH_3 volatilization (Sun et al., 2014). Additionally, it was worthy to note that cumulative NH_3 emissions were slightly higher in soils with the Bm than those with the Bw amendment (Fig. 4 and Table 3c) and that difference could presumably be attributed to less surface area and the much higher pH of Bm (Fig. S3 and Table S1), resulting in weak adsorption and great liming effects. ~~Overall, compared with previous studies (Ro et al., 2015; Mandal et al., 2016), no significant reductions were found in cumulative NH_3 volatilizations over the whole observation period when biochar was added to current vegetable soils. In general, freshly produced biochar typically has very low ability to absorb ammonium (Yao et al., 2012). Over time, biochar surfaces are oxidized and increase adsorption (Wang et al., 2016). Moreover, the recorded increase in CEC by Cheng et al. (2006) indicated that biochars that are sufficiently weathered over a period would increase their ability to retain cations such as NH_4^+–N. Further, relatively long-term experiments are required to elucidate the mechanism and duration of effect.~~

4.2. Biochar effects on vegetable yield and GNrI across different soil types

The application of biochar is usually intended to increase crop yields, and evidence suggests this may be successful (Schulz et al., 2013; Li et al., 2016). Due to its liming effect, biochar helps to improve the supply of essential macro- and micronutrients for plant growth (Chan and Xu, 2009; Major et al., 2010). Enhancement of vegetable yield with bi~~e~~ochar amendment occurred in SD and HLJ soils (Table 3e). Additionally, the effects of Bm and Bw on vegetable yield ~~was~~were ~~mixed~~inconsistent, which probably due to~~ performance of biochars as an amendment is related to~~ the wide diversity of physicochemical characteristics of biochar that translates into variable reactions in soil (Novak et al., 2014). First, compared to Bw, ~~more DOC content was in the~~ Bm has a higher DOC content (Table S1), through which more nutrients may be directly introduced to the soil (Rajkovich et al., 2012). ~~In addition,~~Secondly, besides their large amount of plant-available nutrients (Hass et al., 2012), ~~manure~~ biochars produced with manure have been generally considered significant for improving soil fertility by promoting soil structure development (Joseph et al., 2010), with the result that Bm was found superior to Bw in vegetable production enhancement in our case (Table 3e). As biochar effects on vegetable yield were variable, both biochar properties and soil conditions and crop species ought to be taken into account comprehensively before applying biochar to a certain soil condition.

However, no promotion of yield was observed with biochar amendments in HN and SX. This could be attributed to exacerbated soil salinity, which inhibited the uptake of nutrients and water (Ju et al., 2006; Zhou et al., 2010) and the growth of the soil microorganisms (Setia et al., 2011), leading to unsustainable greenhouse vegetable production. Compared with other biochar (Jia et al., 2012), the higher amounts of ash in Bw and Bm may contain high salts causing soil salinity (Hussain et al., 2016). After the addition of the two salt-rich biochars, the EC values of HN and SX vegetable soils increased and reached the limits to tolerance for the leafy vegetables (Shannon and Grieve, 1998). ~~Additionally, the mixed performance of biochars as an amendment is related to the wide diversity of physicochemical characteristics that translates into variable reactions in soil (Novak et al., 2014). First, compared to Bw, more DOC content was in the Bm (Table S1), through which more nutrients may be directly introduced to the soil (Rajkovich et al., 2012). In addition, besides their large amount of plant-available nutrients (Hass et al., 2012), manure biochars have been generally considered significant for improving soil fertility by promoting soil structure development (Joseph et al., 2010), with the result that Bm was found superior to Bw in vegetable production enhancement (Table 3e). As biochar effects on vegetable yield were variable, both biochar properties and soil conditions and crop species ought to be taken into account comprehensively before applying biochar to a certain soil condition.~~

Here, we assessed two feedstock-derived biochar effects on GNrI in typical cultivated vegetable soils across mainland China. Overall, biochar amendments reduced GNrI over all the soils, with the magnitude largely depending on soil type. Remarkable reduction in GNrI had been detected due to the efficient mitigation induced by biochar in SX and HLJ (Table 3f). However, despite enhanced vegetable yield, no significant decreases in GNrI were observed in SD, mainly because of the absence of mitigation effects on N_2O, NO and NH_3 emissions of biochars (Table 3a, b and c) ~~Additionally~~Overall, ~~divergent influences on GNE and yield were determined with different biochars that~~ Bw was superior to Bm in mitigating the GNrE while Bm performed better in vegetable yield enhancement (Table 3d and e). ~~.~~ Therefore, mitigation efficacys on GNrI were not notably different between Bw and Bm amendments across the four soils.~~,largely due to the divergent influences on GNE and yield that Bw was superior to Bm in mitigating the GNE while Bm performed better in vegetable yield (Table 3d and e). Furthermore, from our perspective, economic effectiveness/feasibility, such as the net ecosystem economic budget, should be considered synchronously in intensive vegetable production before large-scale biochar applicat~~

5. Conclusion

The study demonstrated that biochar amendments mostly~~generally~~ reduced N_2O and NO emissions while slightly increased ~~ithout influencing~~ the NH_3 emissions, while produced no consensus influences on yield though those effects were largely both biochar- and soil-specific. Additionally, biochar amendments did decrease GNrI in intensive vegetable soils across mainland China. Furthermore, Bw was superior to Bm in mitigating the GNrE whereas the Bm performed better in crop yield throughout all soils. Consequently, both soil type and biochar characteristics need to be seriously considered before large-scale biochar application under certain regions of intensive vegetable production.

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References

Ameloot, N., Sleutel, S., Das, K. C., Kanagaratnam, J., and Neve, S. D.: Biochar amendment to soils with contrasting organic matter level: effects on N mineralization and biological soil properties, Global Change Biology Bioenergy, 7, 135–144, 2015.

Anderson, J. and Domsch, K.: A physiological method for the quantitative measurement of microbial biomass in soils, Soil biology and biochemistry, 10, 215–221, 1978.

Anenberg, S. C., Schwartz, J., Shindell, D., Amann, M., Faluvegi, G., Klimont, Z., Janssensmaenhout, G., Pozzoli, L., Van, D. R., and Vignati, E.: Global Air Quality and Health Co-benefits of Mitigating Near-Term Climate Change through Methane and Black Carbon Emission Controls, Environmental Health Perspectives, 120, 831–839, 2012.

Avnery, S., Mauzerall, D. L., Liu, J., and Horowitz, L. W.: Global crop yield reductions due to surface ozone exposure: 1. Year 2000 crop production losses and economic damage, Atmospheric Environment, 45, 2284–2296, 2011.

Behera, S. N., Sharma, M., Aneja, V. P., and Balasubramanian, R.: Ammonia in the atmosphere: a review on emission sources, atmospheric chemistry and deposition on terrestrial bodies, Environmental Science and Pollution Research, 20, 8092–8131, 2013.

Boyer, E. W., Goodale, C. L., Jaworski, N. A., and Howarth, R. W.: Anthropogenic nitrogen sources and relationships to riverine nitrogen export in the northeastern USA. In: The Nitrogen Cycle at Regional to Global Scales, Springer, 2002.

Cayuela, M., Van Zwieten, L., Singh, B., Jeffery, S., Roig, A., and Sánchez-Monedero, M.: Biochar's role in mitigating soil nitrous oxide emissions: A review and meta-analysis, Agriculture, Ecosystems & Environment, 191, 5–16, 2014.

Chan, K. Y. and Xu, Z.: Biochar: nutrient properties and their enhancement, Biochar for environmental management: science and technology, 2009. 67–84, 2009.

~~Cheng, C. H., Lehmann, J., Thies, J. E., Burton, S. D., and Engelhard, M. H.: Oxidation of black carbon by biotic and abiotic processes, Organic Geochemistry, 37, 1477–1488, 2006.~~

Ciais, P.: Carbon and other biogeochemical cycles: Final draft underlying scientific technical assessment, IPCC Secretariat, Geneva, 2013. 2013.

Clough, T. J. and Condron, L. M.: Biochar and the nitrogen cycle: introduction, Journal of Environmental Quality, 39, 1218-1223, 2010.

Clough, T. J., Bertram, J. E., Ray, J. L., Condron, L. M., O'Callaghan, M., Sherlock, R. R., and Wells, N. S.: Unweathered Wood Biochar Impact on Nitrous Oxide Emissions from a Bovine-Urine-Amended Pasture Soil, Soil Science Society of America Journal, 74, 852–860, 2010.

Deng, J., Zhou, Z., Zheng, X., and Li, C.: Modeling impacts of fertilization alternatives on nitrous oxide and nitric oxide emissions from conventional vegetable fields in southeastern China, Atmospheric Environment, 81, 642–650, 2013.

Diao, T., Xie, L., Guo, L., Yan, H., Lin, M., Zhang, H., Lin, J., and Lin, E.: Measurements of N_2O emissions from different vegetable fields on the North China Plain, Atmospheric Environment, 72, 70–76, 2013.

FAO, IIASA, ISRIC, and ISSCAS: Harmonized World Soil Database Version 1.2, 2012. 2012.

Field, J. L., Keske, C. M. H., Birch, G. L., Defoort, M. W., and Cotrufo, M. F.: Distributed biochar and bioenergy coproduction: a regionally specific case study of environmental benefits and economic impacts, Global Change Biology Bioenergy, 5, 177–191, 2013.

Food and Agriculture Organization (FAO) (2015) FAOSTAT (Food and Agriculture Organization Statistical Data) Statistical Yearbook Vol. 4. Available at: http: // faostat.fao.org (accessed 12 August 2015)

Harrison, R. and Webb, J.: A review of the effect of N fertilizer type on gaseous emissions, Advances in Agronomy, 73, 65–108, 2001.

Hass, A., Gonzalez, J. M., Lima, I. M., Godwin, H. W., Halvorson, J. J., and Boyer, D. G.: Chicken manure biochar as liming and nutrient source for acid Appalachian soil, Journal of Environmental Quality, 41, 1096–1106, 2012.

Hu, H. W., Macdonald, C. A., Trivedi, P., Anderson, I. C., Zheng, Y., Holmes, B., Bodrossy, L., Wang, J. T., He, J. Z., and Singh, B. K.: Effects of climate warming and elevated CO_2 on autotrophic nitrification and nitrifiers in dryland ecosystems, Soil Biology & Biochemistry, 92, 1–15, 2016.

Hussain, M., Farooq, M., Nawaz, A., Al-Sadi, A. M., Solaiman, Z. M., Alghamdi, S. S., Ammara, U., Yong, S. O., and Siddique, K. H. M.: Biochar for crop production: potential benefits and risks, Journal of Soils & Sediments, 2016. 1–32, 2016.

IPCC: Climate Change 2013: The Physical Science Basis: working group I contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge University Press, Stockholm, 2013.

~~IPCC, 2013. The Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. New York: Cambridge University Press 1, 535–531.~~

Islam, A., Chen, D., White, R. E., and Weatherley, A. J.: Chemical decomposition and fixation of nitrite in acidic pasture soils and implications for measurement of nitrification, Soil Biology & Biochemistry, 40, 262–265, 2008.

Jia, J., Li, B., Chen, Z., Xie, Z., and Xiong, Z.: Effects of biochar application on vegetable production and emissions of N_2O and CH_4, Soil Science and Plant Nutrition, 58, 503–509, 2012.

Joseph, S. D., Campsarbestain, M., Lin, Y., Munroe, P., Chia, C. H., Hook, J., Van, Z. L., Kimber, S., Cowie, A., and Singh, B. P.: An investigation into the reactions of biochar in soil, Australian Journal of Soil Research, 48, 501–515, 2010.

Ju, X. T., Kou, C. L., Zhang, F. S., and Christie, P.: Nitrogen balance and groundwater nitrate contamination: comparison among three intensive cropping systems on the North China Plain, Environmental Pollution, 143, 117–125, 2006.

Kim, J. Y., Song, C. H., Ghim, Y. S., Won, J. G., Yoon, S. C., Carmichael, G. R., and Woo, J. H.: An investigation on NH_3 emissions and particulate NH_4^+–NO_3^- formation in East Asia, Atmospheric Environment, 40, 2139–2150, 2006.

Kurola, J., Salkinoja-Salonen, M., Aarnio, T., Hultman, J., and Romantschuk, M.: Activity, diversity and population size of ammonia-oxidising bacteria in oil-contaminated landfarming soil, FEMS Microbiology Letters, 250, 33–38, 2005.

Langridge, J. M., Lack, D., Brock, C. A., Bahreini, R., Middlebrook, A. M., Neuman, J. A., Nowak, J. B., Perring, A. E., Schwarz, J. P., and Spackman, J. R.: Evolution of aerosol properties impacting visibility and direct climate forcing in an ammonia-rich urban environment, Journal of Geophysical Research Atmospheres, 117, 2240–2260, 2012.

Li, B., Bi, Z., and Xiong, Z.: Dynamic responses of nitrous oxide emission and nitrogen use efficiency to nitrogen and biochar amendment in an intensified vegetable field in southeastern China, Global Change Biology Bioenergy, 2016. 2016.

Li, B., Fan, C. H., Xiong, Z. Q., Li, Q. L., and Zhang, M.: The combined effects of nitrification inhibitor and biochar incorporation on yield-scaled N_2O emissions from an intensively managed vegetable field in southeastern China, Biogeosciences, 12, 15185–15214, 2015a.

Li, B., Fan, C. H., Zhang, H., Chen, Z. Z., Sun, L. Y., and Xiong, Z. Q.: Combined effects of nitrogen fertilization and biochar on the net global warming potential, greenhouse gas intensity and net ecosystem economic budget in intensive vegetable agriculture in southeastern China, Atmospheric Environment, 100, 10–19, 2015b.

Liu, X., Zhang, A., Ji, C., Joseph, S., Bian, R., Li, L., Pan, G., and Paz-Ferreiro, J.: Biochar's effect on crop productivity and the dependence on experimental conditions—a meta-analysis of literature data, Plant and Soil, 373, 583–594, 2013.

Lu, R.: Methods of soil and agro-chemical analysis, China Agricultural Science and Technology Press, Beijing, 2000. 127–332, 2000. (in Chinese)

Ma, L., Shan, J., Yan, X., 2015. Nitrite behavior accounts for the nitrous oxide peaks following fertilization in a fluvo-aquic soil. Biology and Fertility of Soils 51, 563-572.

Major, J., Lehmann, J., Rondon, M., and Goodale, C.: Fate of soil-applied black carbon: downward migration, leaching and soil respiration, Global Change Biology, 16, 1366–1379, 2010.

~~Mandal, S., Thangarajan, R., Bolan, N. S., Sarkar, B., Khan, N., Ok, Y. S., and Naidu, R.: Biochar-induced concomitant decrease in ammonia volatilization and increase in nitrogen use efficiency by wheat, Chemosphere, 142, 120–127, 2016.~~

Mei, B. L., Zheng, X. H., Xie, B. H., Dong, H. B., Zhou, Z. X., Rui, W., Jia, D., Feng, C., Tong, H. J., and Zhu, J. G.: Nitric oxide emissions from conventional vegetable fields in southeastern China, Atmospheric Environment, 43, 2762–2769, 2009.

Misselbrook, T. H., Weerden, T. J. V. D., Pain, B. F., Jarvis, S. C., Chambers, B. J., Smith, K. A., Phillips, V. R., and Demmers, T. G. M.: Ammonia emission factors for UK agriculture, Atmospheric Environment, 34, 871–880(810), 2000.

Mukherjee, A. and Zimmerman, A. R.: Organic carbon and nutrient release from a range of laboratory-produced biochars and biochar–soil mixtures, Geoderma, s 193–194, 122–130, 2013.

Nelissen, V.: Effect of different biochar and fertilizer types on N_2O and NO emissions, Soil Biology & Biochemistry, 70, 244–255, 2014.

Novak, J. M., Spokas, K. A., Cantrell, K. B., Ro, K. S., Watts, D. W., Glaz, B., Busscher, W. J., and Hunt, P. G.: Effects of biochars and hydrochars produced from lignocellulosic and animal manure on fertility of a Mollisol and Entisol, Soil Use and Management, 30, 175–181, 2014.

Obia, A., Cornelissen, G., Mulder, J., and Dörsch, P.: Effect of Soil pH Increase by Biochar on NO, N_2O and N_2 Production during Denitrification in Acid Soils, Plos One, 10, 359–367, 2015.

Pacholski, A., Cai, G. X., Fan, X. H., Ding, H., Chen, D., Nieder, R., and Roelcke, M.: Comparison of different methods for the measurement of ammonia volatilization after urea application in Henan Province, China, Journal of Plant Nutrition and Soil Science, 171, 361–369, 2008.

Pinder, R. W., Adams, P. J., and Pandis, S. N.: Ammonia emission controls as a cost-effective strategy for reducing atmospheric particulate matter in the Eastern United States, Environmental Science & Technology, 41, 380–386, 2007.

Powlson, D. S., Addiscott, T. M., Benjamin, N., Cassman, K. G., de Kok, T. M., Van, G. H., L'Hirondel, J. L., Avery, A. A., and Van, K. C.: When does nitrate become a risk for humans?, Journal of Environmental Quality, 37, 291–295, 2008.

Rajkovich, S., Enders, A., Hanley, K., Hyland, C., Zimmerman, A. R., and Lehmann, J.: Corn growth and nitrogen nutrition after additions of biochars with varying properties to a temperate soil, Biology & Fertility of Soils, 48, 271–284, 2012.

Ravishankara, A. R., Daniel, J. S., and Portmann, R. W.: Nitrous oxide (N_2O): the dominant ozone-depleting substance emitted in the 21st century, Science, 326, 123–125, 2009.

~~Ro, K. S., Lima, I. M., Reddy, G. B., Jackson, M. A., and Gao, B.: Removing Gaseous NH_3 Using Biochar as an Adsorbent, Agriculture, 5, 991–1002, 2015.~~

Saarnio, S., Heimonen, K., and Kettunen, R.: Biochar addition indirectly affects N_2O emissions via soil moisture and plant N uptake, Soil Biology & Biochemistry, 58, 99–106, 2013.

Schouten, S., Groenigen, J. W. V., Oenema, O., and Cayuela, M. L.: Bioenergy from cattle manure? Implications of anaerobic digestion and subsequent pyrolysis for carbon and nitrogen dynamics in soil, Global Change Biology Bioenergy, 4, 751–760, 2012.

Schulz, H., Dunst, G., and Glaser, B.: Positive effects of composted biochar on plant growth and soil fertility, Agronomy for Sustainable Development, 33, 817–827, 2013.

Setia, R., Marschner, P., Baldock, J., Chittleborough, D., and Verma, V.: Relationships between carbon dioxide emission and soil properties in salt-affected landscapes, Soil Biology & Biochemistry, 43, 667–674, 2011.

Shannon, M. C. and Grieve, C. M.: Tolerance of vegetable crops to salinity, Scientia Horticulturae, 78, 5–38, 1998.

Smith, M. S. and Tiedje, J. M.: Phases of denitrification following oxygen depletion in soil, Soil Biology & Biochemistry, 11, 261–267, 1979.

Smith, P.: Soil carbon sequestration and biochar as negative emission technologies, Global Change Biology, 51, 574–575, 2016.

Sohi, S. P.: Agriculture. Carbon storage with benefits, Science, 338, 1034–1035, 2012.

Sororzano, L.: Determination of ammonia in natural waters by the phenolhypochlorite method, Limnol. Oceanogr, 14, 799–801, 1969.

Spokas, K. A. and Reicosky, D. C.: Impacts of sixteen different biochars on soil greenhouse gas production, Ann. Environ. Sci, 3, 4, 2009.

Stavi, I. and Lal, R.: Agroforestry and biochar to offset climate change: a review, Agronomy for Sustainable Development, 33, 81–96, 2013.

Sun, L., Li, L., Chen, Z., Wang, J., and Xiong, Z.: Combined effects of nitrogen deposition and biochar application on emissions of N_2O, CO_2 and NH_3 from agricultural and forest soils, Soil science and plant nutrition, 60, 254–265, 2014.

Ussiri, D. and Lal, R.: The Role of Nitrous Oxide on Climate Change, Springer Netherlands, 2013.

~~Wang, B., Lehmann, J., Hanley, K., Hestrin, R., and Enders, A.: Ammonium retention by oxidized biochars produced at~~

~~different pyrolysis temperatures and residence times, Rsc Advances, 6, 41907–41913, 2016.~~

Wang, J., Chen, Z., Ma, Y., Sun, L., Xiong, Z., Huang, Q., and Sheng, Q.: Methane and nitrous oxide emissions as affected by organic–inorganic mixed fertilizer from a rice paddy in southeast China, Journal of Soils and Sediments, 13, 1408–1417, 2013.

Wang, J., Chen, Z., Xiong, Z., Chen, C., Xu, X., Zhou, Q., and Kuzyakov, Y.: Effects of biochar amendment on greenhouse gas emissions, net ecosystem carbon budget and properties of an acidic soil under intensive vegetable production, Soil Use and Management, 31, 375–383, 2015a.

Wang, S., Nan, J., Shi, C., Fu, Q., Gao, S., Wang, D., Cui, H., Saizlopez, A., and Zhou, B.: Atmospheric ammonia and its impacts on regional air quality over the megacity of Shanghai, China, Scientific Reports, 5, 2015b.

Wrage, N., Velthof, G., Van Beusichem, M., Oenema, O., 2001. Role of nitrifier denitrification in the production of nitrous oxide. Soil Biology and Biochemistry 33, 1723-1732.

Xiong, Z., Xie, Y., Xing, G., Zhu, Z., and Butenhoff, C.: Measurements of nitrous oxide emissions from vegetable production in China, Atmospheric Environment, 40, 2225–2234, 2006.

~~Yao, Y., Gao, B., Zhang, M., Inyang, M., and Zimmerman, A. R.: Effect of biochar amendment on sorption and leaching of nitrate, ammonium, and phosphate in a sandy soil, Chemosphere, 89, 1467–1471, 2012.~~

Yao, Z., Zheng, X., Xie, B., Mei, B., Wang, R., Klaus, B. B., Zhu, J., and Yin, R.: Tillage and crop residue management significantly affects N-trace gas emissions during the non-rice season of a subtropical rice-wheat rotation, Soil Biology & Biochemistry, 41, 2131–2140, 2009.

Zhang, A., Cui, L., Pan, G., Li, L., Hussain, Q., Zhang, X., Zheng, J., and Crowley, D.: Effect of biochar amendment on yield and methane and nitrous oxide emissions from a rice paddy from Tai Lake plain, China, Agriculture Ecosystems & Environment, 139, 469–475, 2010.

Zhang, F., Chen, X., and Vitousek, P.: Chinese agriculture: An experiment for the world, Nature, 497, 33-35, 2013.

Zhang, M., Fan, C. H., Li, Q. L., Li, B., Zhu, Y. Y., and Xiong, Z. Q.: A 2-yr field assessment of the effects of chemical and biological nitrification inhibitors on nitrous oxide emissions and nitrogen use efficiency in an intensively managed vegetable cropping system, Agriculture Ecosystems & Environment, 201, 43–50, 2015.

Zhang, Y., Luan, S., Chen, L., and Shao, M.: Estimating the volatilization of ammonia from synthetic nitrogenous fertilizers used in China, Journal of Environmental Management, 92, 480–493, 2011.

Zhao, L. M., Wu, L. H., Dong, C. J., and Li, Y. S.: Rice yield, nitrogen utilization and ammonia volatilization as influenced by modified rice cultivation at varying nitrogen rates, Agricultural Sciences, 01, 10–16, 2010.

Zhao, X., Wang, J., Wang, S., and Xing, G.: Successive straw biochar application as a strategy to sequester carbon and

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improve fertility: A pot experiment with two rice/wheat rotations in paddy soil, Plant and Soil, 378, 279–294, 2014.

Zheng, X., Mei, B., Wang, Y., Xie, B., Wang, Y., Dong, H., Xu, H., Chen, G., Cai, Z., and Yue, J.: Quantification of N_2O fluxes from soil–plant systems may be biased by the applied gas chromatograph methodology, Plant and Soil, 311, 211–234, 2008.

Zhou, J. B., Chen, Z. J., Liu, X. J., Zhai, B. N., and Powlson, D. S.: Nitrate accumulation in soil profiles under seasonally open ‘sunlight greenhouses’ in northwest China and potential for leaching loss during summer fallow, Soil Use and Management, 26, 332–339, 2010.

Zhu, T., Zhang, J., Cai, Z., 2011. The contribution of nitrogen transformation processes to total N_2O emissions from soils used for intensive vegetable cultivation. Plant and Soil 343, 313-327.

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Table legends

Table 1

Soil organic carbon (SOC), soil total nitrogen (TN), soil pH, electric conductivity (EC) and microbial biomass carbon (MBC) as affected by different treatments across the four vegetable soils.

Soil	Treatment	SOC (g kg^{-1})	TN (g kg^{-1})	pH	EC (ds m^{-1})	MBC (mg kg^{-1})
HN	N	8.0 ±0.8c	1.37 ±0.12b	4.37 ±0.04c	1.76 ±0.21b	1353 ±119a
	N+Bw	15.6 ±0.5b	1.47 ±0.07b	4.64 ±0.04b	2.43 ±0.31a	1173 ±49b
	N+Bm	18.8 ±0.6a	1.64 ±0.04a	5.01 ±0.03a	2.00 ±0.32ab	1234 ±50ab
SX	N	9.7 ±0.7c	1.55 ±0.04b	7.53 ±0.02b	1.74 ±0.27b	490 ±9a
	N+Bw	15.6 ±0.8b	1.62 ±0.06b	7.61 ±0.05a	2.25 ±0.22a	495 ±16a
	N+Bm	17.5 ±1.1a	1.79 ±0.03a	7.63 ±0.01a	1.96 ±0.06ab	504 ±18a
SD	N	7.9 ±0.1b	1.13 ±0.04b	7.70 ±0.08a	0.85 ±0.03b	535 ±13b
	N+Bw	14.2 ±0.6a	1.20 ±0.04b	7.66 ±0.03a	0.92 ±0.04a	554 ±10ab
	N+Bm	15.5 ±1.4a	1.37 ±0.06a	7.71 ±0.03a	0.87 ±0.02ab	573 ±12a
HLJ	N	29.9 ±0.5b	2.19 ±0.04b	6.91 ±0.05a	0.83 ±0.03b	921 ±44b
	N+Bw	36.0 ±1.5a	2.20 ±0.03b	6.92 ±0.06a	0.95 ±0.03a	988 ±56b
	N+Bm	38.1 ±1.8a	2.41 ±0.01a	6.94 ±0.04a	0.92 ±0.06a	1242 ±196a
ANOVA results						
Biochar		***	***	***	***	*
Soil		***	***	***	***	***
Biochar ×Soil		*	n.s.	***	n.s.	**

Data shown are means ± standard deviations of three replicates. See Fig. 1 for treatments codes. Different letters within the same column indicate significant differences among treatments within the same soil at $p < 0.05$ level.

***Significant at $p < 0.001$; **significant at $p < 0.01$; *significant at $p < 0.05$; n.s. not significant.

Table 2

Two-way ANOVA and mean effects of biochar (Bc) and soil (S) types on cumulative ~~gaseous nitrogen (~~N_2O, NO and NH_3~~)~~ emissions, gaseous reactive nitrogen emission (GNrE), vegetable yield and gaseous reactive nitrogen intensity (GNrI) during the entire sampling period.

Factors	DF	N_2O emission			NO emission			NH_3 emission			GNrE			Vegetable yield			GNrI		
		SS	F	P	SS	F	P	SS	F	P	SS	F	P	SS	F	P	SS	F	P
Bc	2	271.9	65.1	***	46.4	174.7	***	0.5	0.8	n.s.	380.5	86.4	***	76.2	3.2	n.s.	0.1	7.9	**
S	3	1429.9	228.1	***	152.2	382.1	***	4.1	3.8	*	2322.6	351.5	***	4316.9	123.3	***	2.3	110.3	***
Bc×S	6	179.3	14.3	***	33.4	41.9	***	1.4	0.7	n.s.	234.5	17.7	***	230.4	3.3	*	0.1	1.6	n.s.
Model	11	4009.7	174.5	***	225.3	154.3	***	29.1	7.5	***	5290	218.3	***	15962.0	124.4	***	5.8	77.0	***
Error	24	50.1			3.2			8.5			52.9			280.0			0.2		
biochar effect (n = 9)																			
N mean		12.01 ± 1.44a			2.86 ± 0.24a			5.92 ± 0.24b			43.81 ± 1.25b			20.50 ± 1.60a			0.57 ± 0.05a		
N+Bw mean		7.01 ± 0.58b			1.55 ± 0.14b			6.65 ± 0.27a			43.53 ± 1.67b			14.94 ± 0.84b			0.45 ± 0.04b		
N+Bm mean		10.37 ± 0.56a			1.55 ± 0.10b			7.01 ± 0.25a			49.53 ± 1.11a			18.60 ± 0.65a			0.49 ± 0.03ab		
Soil effect (n = 9)																			
HN mean		27.20 ± 1.85a			5.80 ± 0.50a			5.31 ± 0.16c			33.06 ± 1.65c			38.04 ± 1.90a			1.15 ± 0.11a		
SX mean		4.89 ± 0.45b			1.08 ± 0.13b			12.69 ± 0.46a			25.05 ± 1.11d			12.69 ± 0.46b			0.51 ± 0.01b		
SD mean		2.25 ± 0.26c			0.25 ± 0.09c			9.51 ± 0.55b			44.88 ± 0.49b			9.51 ± 0.55c			0.21 ± 0.01c		
HLJ mean		4.48 ± 0.68b			0.81 ± 0.04b			11.79 ± 0.71a			79.50 ± 2.41a			11.79 ± 0.71b			0.15 ± 0.01c		

带格式表格

SS: the sum of squares.

F value: the ratio of mean squares of two independents samples.

P value: the index of differences between the control group and the experimental group. *, ** and *** indicate significance at $p < 0.05$, $p < 0.01$ and $p < 0.001$, respectively.

n.s.: not significant.

Data shown are means ± standard deviations of the nine replicates. See Fig. 1 for treatments codes. Different letters within the same column indicate significant differences among treatments at $p < 0.05$ level.

Table 3

Cumulative gaseous nitrogen (N_2O, NO and NH_3) emissions, gaseous reactive nitrogen emission (GNrE), vegetable yield and gaseous reactive nitrogen intensity (GNrI) under the different treatments across the four soils.

Treatments	HN	SX	SD	HLJ
(a) Cumulative N_2O emissions (kg N ha^{-1})				
N	30.59 ±3.15a	7.83 ±0.60a	2.52 ±0.37a	7.10 ±1.91a
N+Bw	19.45 ±2.43b	3.20 ±0.28b	1.97 ±0.21a	3.45 ±0.86b
N+Bm	31.56 ±1.35a	3.63 ±0.62b	2.26 ±0.58a	4.01 ±0.68b
(b) Cumulative NO emissions (kg N ha^{-1})				
N	8.99 ±1.01a	1.27 ±0.15a	0.20 ±0.08a	0.97 ±0.11a
N+Bw	4.54 ±0.60b	0.80 ±0.13b	0.33 ±0.19a	0.52 ±0.03b
N+Bm	3.87 ±0.30b	1.16 ±0.17a	0.21 ±0.10a	0.94 ±0.03a
(c) Cumulative NH_3 emissions (kg N ha^{-1})				
N	4.72 ±0.27a	5.79 ±0.54b	6.34 ±0.51a	5.67 ±0.42a
N+Bw	5.09 ±0.38a	6.83 ±0.74ab	7.35 ±0.75a	6.24 ±0.49a
N+Bm	5.32 ±0.42a	7.57 ±0.57a	7.37 ±1.11a	6.48 ±0.43a
(d) GNrE (kg N ha^{-1})				
N	44.30 ±3.13a	14.89 ±1.33a	9.06 ±0.80a	13.74 ±1.67a
N+Bw	29.08 ±2.21b	10.82 ±1.14b	9.64 ±0.88a	10.21 ±0.92b
N+Bm	40.76 ±1.66a	12.36 ±0.74b	9.84 ±0.49a	11.42 ±0.27b
(e) Vegetable yield (t ha^{-1})				
N	35.20 ±2.52a	25.29 ±3.90a	39.09 ±2.03b	75.65 ±5.84b
N+Bw	29.05 ±2.35b	23.57 ±1.74a	44.53 ±3.74b	76.95 ±4.04ab
N+Bm	34.93 ±2.87a	26.30 ±2.63a	51.00 ±3.18a	85.89 ±3.29a
(f) GNrI (kg N t^{-1} yield)				
N	1.27 ±0.18a	0.59 ±0.08a	0.23 ±0.02a	0.18 ±0.04a
N+Bw	1.01 ±0.12a	0.46 ±0.05b	0.22 ±0.04a	0.13 ±0.02b
N+Bm	1.17 ±0.15a	0.47 ±0.04b	0.19 ±0.01a	0.13 ±0.01b

Data shown are means ± standard deviations of the three replicates. See Fig. 1 for treatments codes. Different letters within the same column indicate significant differences among treatments within the same soil at $p < 0.05$ level.

Table 4

The correlations between N_2O or NO emission and PNR or DEA in each soil.

Item	HN		SX		SD		HLJ	
	PNR	DEA	PNR	DEA	PNR	DEA	PNR	DEA
N_2O	0.75*	0.66	0.49	0.76*	-0.10	0.16	-0.82**	0.70*
NO	0.62	-0.29	0.79*	0.69*	-0.54	0.01	-0.63	0.22

Asterisks indicated 0.05 level significances (*$p < 0.05$) and 0.01 level significances (**$p < 0.01$), n = 9.

带格式表格

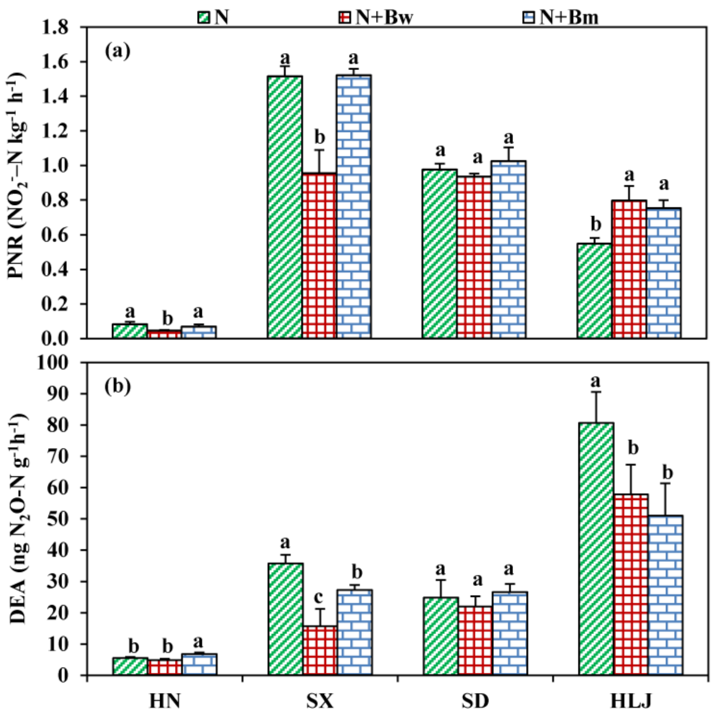
Figure legends

Fig. 1 Potential nitrification rate (PNR) and Denitrification enzyme activity (DEA) under different treatments in HN, SX, SD and HLJ soils. The three treatments with each soil were urea without biochar (N), urea with wheat straw biochar (N+Bw) and urea with swine manure biochar (N+Bm). Bars indicate standard deviation (mean + SD, n = 3). Different letters above the bars indicate significant differences among the different treatments within the same soil, at $p < 0.05$.

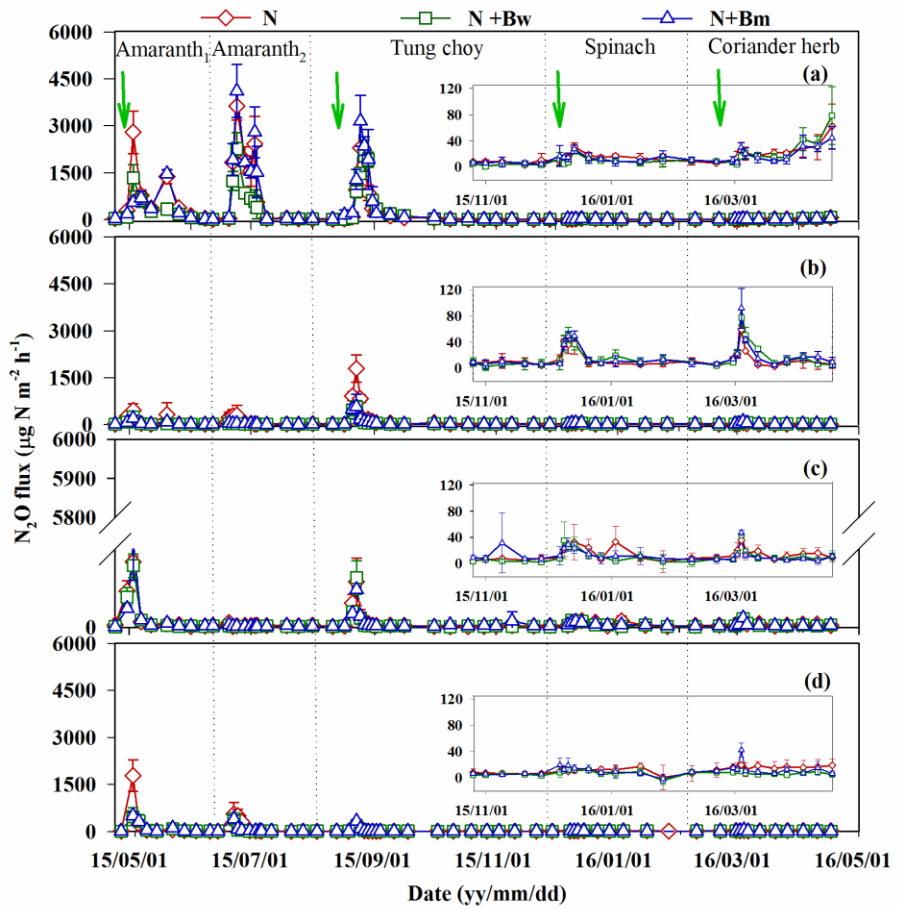
Fig. 2 Temporal dynamics of soil N_2O ($\mu g\ N\ m^{-2}\ h^{-1} \pm SD$, n = 3) fluxes under different treatments in HN (a), SX (b), SD (c) and HLJ (d) vegetable soils with five consecutive vegetable crops. The solid arrows indicate fertilization. See Fig. 1 for treatments codes.

Fig. 3 Temporal dynamics of soil NO ($\mu g\ N\ m^{-2}\ h^{-1} \pm SD$, n = 3) fluxes under different treatments in HN (a), SX (b), SD (c) and HLJ (d) vegetable soils with five consecutive vegetable crops. The solid arrows indicate fertilization. See Fig. 1 for treatments codes.

Fig. 4 Cumulative ammonia (NH_3) emissions from the HN (a), SX (b), SD (c) and HLJ (d) soils during the four nitrogen fertilization events F: every N fertilization event. The bars indicate the standard deviation of the mean (kg N $ha^{-1} \pm SD$, n = 3) of each treatment for the sum of the four N fertilization events. See Fig. 1 for treatments codes. Different letters above the bars indicate significant differences among the different treatments for each soil, at $p < 0.05$.

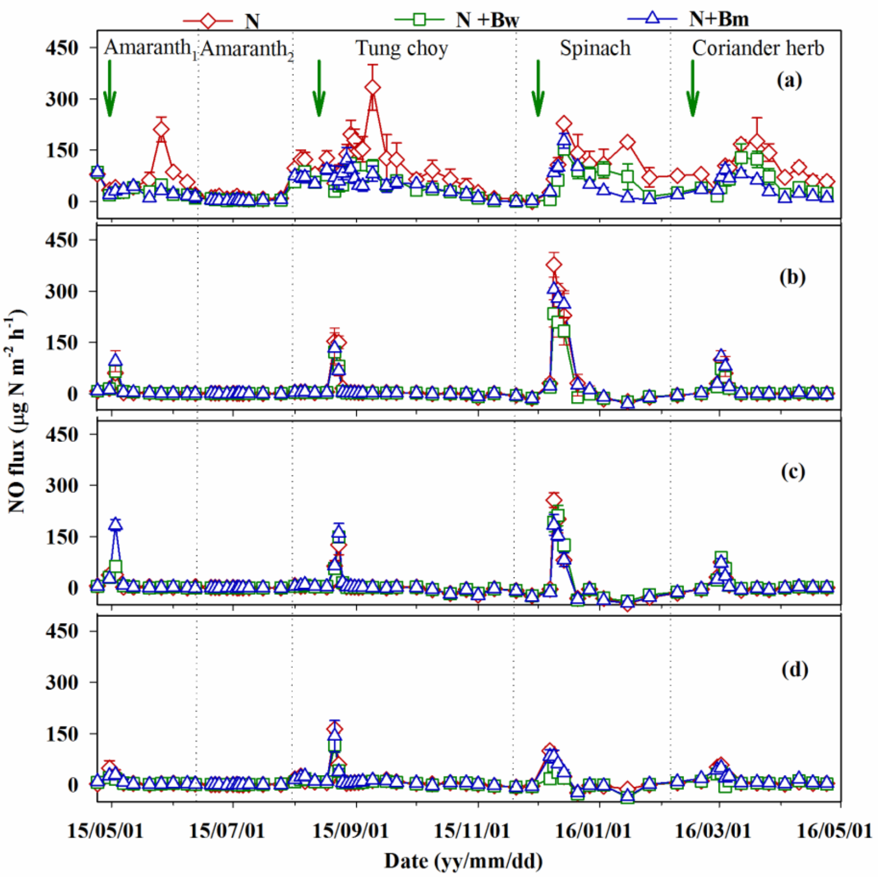

N
N+Bw
N+Bm
(a)
PNR (NO_2^--N kg^{-1} h^{-1})
(b)
DEA (ng N_2O-N g^{-1}h^{-1})
HN
SX
SD
HLJ

1

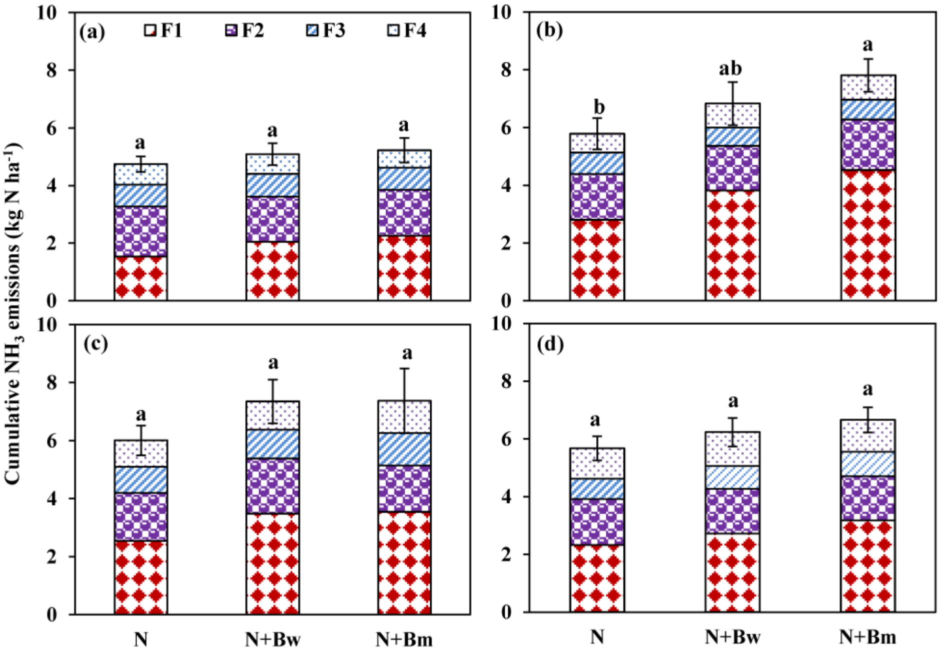


N
N +Bw
N+Bm
Amaranth1
Amaranth2
Tung choy
Spinach
Coriander herb
(a)
(b)
(c)
(d)
N2O flux (μg N m-2 h-1)
6000
4500
3000
1500
0
5900
5800
120
80
40
15/11/01
16/01/01
16/03/01
15/05/01
15/07/01
15/09/01
16/05/01
Date (yy/mm/dd)

1


N
N +Bw
N+Bm
Amaranth1
Amaranth2
Tung choy
Spinach
Coriander herb
(a)
(b)
(c)
(d)
450
300
150
0
NO flux (μg N m-2 h-1)
15/05/01
15/07/01
15/09/01
15/11/01
16/01/01
16/03/01
16/05/01
Date (yy/mm/dd)

1



(a)
F1
F2
F3
F4
(b)
(c)
(d)
Cumulative NH_3 emissions (kg N ha^{-1})
0
2
4
6
8
10
a
b
ab
N
N+Bw
N+Bm