



Characterization and influence of biochars on nitrous oxide emission from agricultural soil

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ABSTRACT

Extensive use of biochar to mitigate N₂O emission is limited by the lack of understanding on the exact mechanisms altering N₂O emissions from biochar-amended soils. Biochars produced from giant reed were characterized and used to investigate their influence on N₂O emission. Responses of N₂O emission varied with pyrolysis temperature, and the reduction order of N₂O emission by biochar (BC) was: BC200 ≈ BC600 > BC500 ≈ BC300 ≈ BC350 > BC400. The reduced emission was attributed to enhanced N immobilization and decreased denitrification in the biochar-amended soils. The remaining polycyclic aromatic hydrocarbons (PAHs) in low-temperature biochars (300–400 °C) played a major role in reducing N₂O emission, but not for high-temperature biochars (500–600 °C). Removal of phenolic compounds from low-temperature (200–400 °C) biochars resulted in a surprising reduction of N₂O emission, but the mechanism is still unknown. Overall, adding giant reed biochars could reduce N₂O evolution from agricultural soil, thus possibly mitigating global warming.

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1. Introduction

Nitrous oxide (N₂O) has received great attention because it is a powerful greenhouse gas (GHGs) with its current concentration of 319 ppb in the atmosphere, which can persist for up to 150 years and has a global warming potential 298 times that of CO₂ (IPCC, 2007). It is also the dominant source of ozone-depleting nitrogen oxides in the stratosphere (Crutzen, 1970). The foremost source of anthropogenic N₂O is agriculture, which has been stimulated by the use of synthetic nitrogen fertilizers for food production (Davidson, 2009). The surplus N with low use efficiency by crops was susceptible to loss as N₂O emission (Bhatia et al., 2010). Hence, small reductions in N₂O emissions could potentially contribute significant benefits for environmental protection and agricultural production. Practices that reduce soil N₂O emission include using slow or controlled release fertilizer, nitrification inhibitors, comprehensive management of water and fertilizer, maize straw return without burning into field, reduced or no tillage (Guo and Zhou, 2007; Bhatia et al., 2010; Jiang et al., 2010). Although these technologies may mitigate N₂O emission to some extent, they also have their own shortcomings. For example, application of slow or controlled release fertilizer can increase agricultural cost and nitrification inhibitors addition may cause

secondary input pollution of soils. As a major N fertilizer producer and user, China is urged to reduce GHGs emissions from agricultural soil to meet the target of total reduction of GHGs emission by 40–45% per unit of GDP by 2020 (Zhang et al., 2010). Therefore, new highly efficient and environmental friendly products have become obligatory to develop for mitigating N₂O emission from agricultural soils.

Biochar, a carbon-rich material, produced from biomass under partial or complete exclusion of oxygen and used as a soil amendment, may be a potential solution (Woolf et al., 2010). Biochar application for improving soil quality and carbon sequestration has generated great interest for scientists and policy makers (Spokas et al., 2011a). Some research to date shows that biochar has the potential to manipulate the N cycling rates in soils via altering the net nitrification rate (Deluca et al., 2006), stimulating N immobilization (Rondon et al., 2007), increasing NH₄⁺-N storage (Taghizadeh-Toosi et al., 2012), decreasing NH₃ volatilization (Steiner et al., 2010), enhancing ammonium-oxidizing bacterial abundance (Ball et al., 2010) and improving N availability for crops (Rondon et al., 2007). Its influence on these processes may further implicate that biochar may mitigate the N₂O emission from agricultural soils. A few research reported that incorporation of biochar effectively reduced N₂O emission from different soils with or without N fertilizer additions (Rondon et al., 2005; Spokas et al., 2009; Zhang et al., 2010). For example, Rondon et al. (2005) reported that biochar decreased N₂O emissions by 50% and 80% under soybean and grass systems,

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respectively. However, the suppression effects are not always consistent, with N_2O being found to increase or not altered in some biochar treated trials. Clough et al. (2010) reported that fluxes of N_2O from the biochar plus urine treatment were generally higher than from urine alone during the first 30 d, but after 50 d there was no significant difference. Scheer et al. (2011) also documented that the cattle feedlot waste biochar had no significant effect on N_2O emission from red Ferrosol. The varied effects of biochars on N_2O emission and the exact mechanisms underlying the mitigation of N_2O emission from biochar-amended soils were still not completely clear, which may be caused by the utilization of a great variety of soils and biochars (Yoo and Kang, 2012). Thus, more research should be focused on the effects of specific types of biochar on N_2O emission from specific soils to further identify the exact mechanisms underlying the response of N_2O emission from biochar-amended soil.

The potential explanations for the N_2O mitigation have mainly focused on abiotic interactions in the biochar-amended soils such as changes of pH, decrease of bulk density and water penetration, improvement of soil structure and nutrients availability and increase of sorption capacity (e.g., NH_4^+ , NH_3) (Spokas et al., 2009; Singh et al., 2010; Beesley et al., 2011; Taghizadeh-Toosi et al., 2011, 2012). The abiotic interaction induced by biochar may alter biotic processes in soils, which include nitrification and denitrification, being the main pathway of N_2O formation. However, the effects of biochar on soil biota especially nitrobacteria and denitrifying bacteria have received much less attention than its effects on soil abiotic interactions (Ball et al., 2010). The effects of biochar on soil abiotic and biotic interactions would be largely dependent on the physical and chemical properties of biochar. Several studies reported that organic compounds such as phenolic compounds (PHCs) (Karagöz et al., 2005), polycyclic aromatic hydrocarbons (PAHs) and dioxins (McGrath et al., 2003; Hale et al., 2012; Hilber et al., 2012) would be formed during biochar production. The formation of these compounds retained in biochars is affected by the types of feedstock and pyrolysis temperature (Hale et al., 2012). It was reported that phenolic compounds (PHCs) were formed at lower temperatures (e.g., 280 °C) (Karagöz et al., 2005), while PAHs were formed at relatively higher temperature (e.g., >350 °C) (McGrath et al., 2003; Hilber et al., 2012). These compounds can be present in the biochar matrix and even bioavailable and toxic to exposed organisms (Lee et al., 2003; Berglund et al., 2004). Thus, it is reasonable to hypothesize that low-temperature biochar (<300 °C) may suppress the soil N_2O emission mainly due to the toxicity of PHCs to denitrifying bacteria; but relatively high-temperature biochar (>350 °C) due to the PAHs toxicity. There have, to our knowledge, been no studies performed on the effects of biochar and its contained PHCs and PAHs on soil N_2O emission.

Giant reed (*Arundo donax* L.) (GR), a perennial herb, is widely used in the process of reverting cultivated land to wetland in China (Li et al., 2011). It could be an ideal material for biochar production due to its fast growth and high yield. Thus, a series of biochars were produced at different temperatures from giant reed stems without leaves and the physical and chemical properties were also characterized. Laboratory experiments were conducted with these biochars to assess the influence of the biochar addition into an agricultural soil on N_2O emissions and identify the possible roles of PAHs and PHCs in these biochars for the effects of biochar on N_2O emissions.

2. Materials and methods

2.1. Soil

Surface soil (0–20 cm) was collected from a cropland (120.38S, 36.29E) after potato was harvested at Chenyang district of Qingdao, Shandong province, China. The soil was air-dried and ground to pass through a 2 mm sieve and thoroughly

homogenized. The soil is classified as silt clay. The physical and chemical properties of the soil are: pH 6.64 (1:2.5 w/v ratio H_2O), total N 0.72 g kg^{-1} , NH_4^+-N 6.03 mg kg^{-1} , NO_3^-+N 12.42 mg kg^{-1} , total P 483 mg kg^{-1} , total organic carbon 9.50 g kg^{-1} , sand 19.2%, silt 45.7% and clay 35.1%.

2.2. Biochar preparation

Biochars were produced using slow pyrolysis. Briefly, GR stems without leaves were pyrolyzed at 200, 300, 350, 400, 500 or 600 °C for 2 h using a vacuum tube furnace (O-KTF1200, Chunlei Co., China) under a N_2 flow of 500 mL min^{-1} . The prepared biochars were hereafter referred to as BC200, BC300, BC350, BC400, BC500 and BC600, respectively. Biochar samples were milled to pass a 0.2 mm sieve prior to further analysis. For comparison, raw GR material (BC0) was included in this study.

To remove PHCs remained in biochars, 1 g of BC200, BC300, BC350 or BC400 was rinsed with 25 ml 50% methanol overnight and then filtered, respectively (Gundale and DeLuca, 2006). The procedure was repeated four times. Then the biochar samples were dried at 75–80 °C for 72 h. The biochar samples without PHCs are hereafter referred to as BC200-P, BC300-P, BC350-P and BC400-P, respectively. The PAHs contained in BC350, BC400, BC500 and BC600 were removed by the same procedure except using acetone/hexane solvent (1:1 v/v) (Gomez-Eyles et al., 2010). The PAHs-free biochar samples are hereafter referred to as BC350-A, BC400-A, BC500-A and BC600-A, respectively. Additionally, a portion of BC350-P and BC400-P was also washed as stated above to further remove the contained PAHs, hereafter referred to as BC350-P-A and BC400-P-A.

2.3. Biochar characterization

Total C, N, H, and O were determined in duplicate with an elemental analyzer (MicroCube, Elementar, Germany). Dissolved organic carbon (DOC) was measured by a TOC analyzer (TOC-L, Shimadzu, Japan). Ash content was measured by heating the biochars at 750 °C for 4 h. Fourier transform infrared (FTIR) absorbance spectra of KBr pellets prepared with 0.5% biochar were recorded using a Perkin–Elmer spectrometer (Spectrum One, Perkin–Elmer, USA) with a diffuse-reflectance sampling accessory between 450 and 4000 cm^{-1} with one hundred scans averaged with a resolution of 4 cm^{-1} . Surface area was analyzed using Quantachrome Autosorb-1 (Autosorb-1, Quantachrome, USA). The Brunauer–Emmett–Teller (BET) surface areas (S_{BET}) were obtained from N_2 adsorption at 77 K. The cumulative surface area (S_{CSA}) up to 1.4 nm was determined from CO_2 isotherms at 273 K between 1×10^{-6} and 0.03 relative pressure using grand canonical Monte Carlo simulation and density functional theory calculations with built-in software (Autosorb 1.55, Quantachrome, USA).

Water-soluble and total phenolic compounds (WPHCs and TPHCs) were measured by extracting 1 g of ground biochar in 25 ml of deionized water and 50% methanol, respectively, and were analyzed using the Prussian blue method (Gundale and DeLuca, 2006). The total PAHs contained in biochars were extracted via Soxhlet extractions (2 g biochar, 60 °C, 24 h). *N*-hexane and dichloromethane (1:1, v:v) were used as the extraction solvent as it has previously been identified to give optimal recovery of PAHs from charcoals (Hilber et al., 2012). Then the extracts were condensed to about 2 mL via rotary evaporation, and enriched with solid phase extraction column (Flory silicon, capacity 1 g, CNW). The filtrate was then purged to 1 mL with high purity nitrogen. The PAHs concentration in the extract was analyzed by GC–MS (7890-5975, Agilent, USA) equipped with an ion trap detector using an Agilent DB-5 column. The oven was temperature-programmed from 80 to 250 °C at 10 °C min^{-1} for 3 min, from 250 to 280 °C at 8 °C min^{-1} for 7 min, from 280 to 300 °C at 3 °C min^{-1} for 2 min.

2.4. N_2O emission and sampling

The experiment was carried out with 60 g soil per replicate (five replicates per treatment), incubated in a 1000 mL wide-mouth glass bottle (Fig. S1). Biochar was added at a rate of 5% (w/w), mixed thoroughly in the bottle. Then 23.8 mL distilled water contained 7.28 g L^{-1} KNO_3 (equivalent to 400 mg N kg^{-1} soil) was uniformly dripped on the soil. All the incubations were carried out at the maximum water holding capacity (WHC), at which the N_2O was significantly produced in the soil (Fig. S2). The bottles were incubated at 30 °C in dark throughout the incubation. Gas samples were taken periodically during the incubation. Before gas sampling, the bottle headspace was fanned gently for 10 s and a gas sample was withdrawn from the open headspace above the soil, this represented the “zero minute” sampling time. Subsequently, gas samples were withdrawn from the enclosed headspace at 40 min after the bottles sealed with rubber stopper pre-fitted with 2 mm tube topped with a three-way stopcock, to which a 20 mL syringe was attached for gas sampling (Fig. S1). N_2O production flux was determined from the linear increase in headspace concentrations over the incubation period (Spokas et al., 2009; Singh et al., 2010).

2.5. Gaseous and soil sample analysis

N_2O concentration in the collected gas sample was analyzed using a gas chromatograph (SHIMADZU GC-14B, Kyoto, Japan) equipped with an electron capture

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