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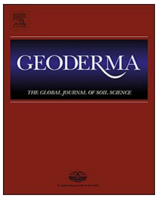

Leaching of ammonium and nitrate from Acrisol and Calcisol amended with holm oak biochar: A column study

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ABSTRACT

Biochar is a carbon-rich porous material intensively studied for its agronomic benefits, such as decrease of greenhouse gases emission and nutrient losses via leaching, increased crop production and improved soil physical and chemical properties. We investigated the effect of holm oak biochar produced at 600 °C on mineral nitrogen (N) leaching from two contrasting soils (Acrisol and Calcisol). Biochar was applied at three rates (0%, 1% and 2% w/w) with (B0-F, B1-F and B2-F, respectively) and without (B0, B1 and B2, respectively) ammonium-based fertilizer. Soil columns were leached with deionized water and mineral N in leachate was monitored during ten weeks after each fertilization. Sorption behavior of biochar-amended soils was assessed in batch experiments before and after leaching. Biochar increased ammonium (NH_4^+-N) sorption in sandy Acrisol but had no effect on nitrate (NO_3^- -N) sorption. Furthermore, sorption properties of soil decreased by up to 25% during the study. In Acrisol, biochar affected NH_4^+-N leaching, which was increased by both doses of biochar without fertilization, but decreased by the lower biochar application rate when fertilizer was added. The leaching of NO_3^--N was not affected by biochar in Acrisol. The ability of Calcisol to adsorb NH_4^+-N was high and was not further increased by biochar, which corresponds to no NH_4^+-N leaching from Calcisol regardless the biochar application rate. Moreover, biochar had no effect on NO_3^--N leaching from Calcisol. Our results demonstrate that biochar effect on leaching of inorganic N forms is inconsistent, evolves in time and is highly dependent on soil properties. Alleviation of soil acidity by biochar application to Acrisol resulted in short-term stimulation organic N mineralization, which resulted in enhanced amount of NH_4^+-N being leached. Furthermore, the fact that biochar lost its effect on N leaching already after the second fertilizer application could cast a doubt on the efficiency of biochar application to soil in order to increase N retention and decrease N leaching.

1. Introduction

Biochar, a carbon-rich material obtained by pyrolysis of organic matter, has been a focus of wide attention due to its potential to mitigate climate change via carbon (C) sequestration (Lehmann et al., 2006) and reduction of greenhouse gas emissions (Case et al., 2015; Cayuela et al., 2013). There is an exponentially growing number of studies focusing on the agronomic benefits of biochar including positive effects on soil properties (Mukherjee et al., 2014) and crop growth (Jeffery et al., 2011; Wang et al., 2012). Although biochar can contain significant amount of nutrients, from the practical or economical point of view it is usually not suitable for annual application to the soil. Instead, its potential lies in the fact that it can serve as a slow-release fertilizer liberating the nutrients continuously and thus, preventing the losses caused by the excess of available and mobile nutrients when there is no crop demand. Despite the large scale of potential modes of biochar's effects on nitrogen (N) cycling (Clough et al., 2013), the interactions of these mechanisms and the interplay between soil-

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microorganisms-biochar system remain poorly understood. Biochar has been observed to affect N fixation (Mia et al., 2014), N mineralization or immobilization (Ameloot et al., 2015), to increase or decrease nitrification (Ulyett et al., 2014), denitrification (Cayuela et al., 2013), or ammonia volatilization (Mandal et al., 2016).

Soil degradation, contamination of ground water and the high cost of fertilizers all lead to an urgent need of mechanisms to improve nutrient retention in soil and to prevent nutrient losses. Due to its porous structure and surface charge, biochar is a promising tool for N leaching mitigation (Laird et al., 2010). Clough et al. (2013) reviewed the effects of biochar on N dynamics and summarized the potential mechanisms of reduced N leaching to (i) adsorption onto biochar surface, (ii) anion or cation exchange reactions and (iii) immobilization as a result of the addition of a labile C contained in biochar. Increasing number of studies demonstrate that biochar could decrease N leaching due to its large surface area and surface charge (Ding et al., 2010; Yao et al., 2012). Indeed, its cation exchange capacity (CEC) is most likely the reason for ammonium (NH_4^+-N) retention by biochar (Dempster et al., 2011). Nevertheless, the sorption properties of biochar depend on feedstock and pyrolysis temperature. For example, Yao et al. (2012) reported that biochars produced at temperatures of 600 °C or higher displayed the highest nitrate (NO_3^--N) adsorption. Furthermore, also soil properties will affect the final sorption behavior of the soil-biochar mixture (Streubel et al., 2011).

During the pyrolysis, the major part of the biomass C is transformed and becomes recalcitrant, leaving only a minor part available to microorganisms (Wang et al., 2015). This small C pool can lead to a short-term stimulation of microbial processes, N immobilization (Zavalloni et al., 2011) or N-mining resulting in increased soil organic matter (SOM) mineralization. Indeed, increased (Castaldi et al., 2011), decreased (Dempster et al., 2011) and unaffected (Castaldi et al., 2011; Streubel et al., 2011) net N mineralization have been reported. Although both biotic and abiotic degradation of biochar have been demonstrated to have an effect on biochar surface properties and sorption capacity (Hale et al., 2011; Liu et al., 2013), most studies focus on sorption behavior of fresh biochar and fewer studies deal with aged or degraded biochar (Gronwald et al., 2015; Liu et al., 2013). Furthermore, the stage of oxidation of biochar could affect N transformation processes. For instance, nitrification rates were observed to increase after charcoal application (Berglund et al., 2004; DeLuca et al., 2006) but also decreased when fresh biochar was used (Zheng et al., 2012).

Growing number of studies aims to clarify the mechanism involved in biochar-induced reduction of leaching of highly mobile NO_3^--N from soil profile (Kanthle et al., 2016). The application of biochar to soil may affect the fate of applied fertilizer by various mechanisms, such as direct sorption of NO_3^--N (Mukherjee et al., 2014) which retains NO_3^--N in soil for a longer period of time increasing the opportunities of NO_3^--N uptake by plants or soil microorganisms; or by adsorption of NH_4^+-N which can prevent nitrification (Liang et al., 2006). Biochar sorption could limit NH_4^+-N assimilation by soil microorganisms or plants or its use as energy source of ammonia oxidizers, processes that need NH_4^+-N in solution (Thies et al., 2015).

Apart from biochar feedstock and production conditions, also soil type determines the properties of soil-biochar mixtures and the final agronomic or environmental impact of biochar application to soil (Streubel et al., 2011). Soil N transformations are likely to be affected distinctly in different soil types (Clough et al., 2013). While biochar porosity and its positive effect on soil aggregation (Herath et al., 2013) could be of higher interest in poorly drained soils, biochar sorption potential and high CEC could be a promising mean to lower nutrient losses in sandy (Sika and Hardie, 2014) or kaolinitic soils (Laird et al., 2010). Furthermore, the native SOM content has been observed to affect the role of biochar in NO_3^--N leaching mitigation (Kanthle et al., 2016). Besides soil C and N content, the recalcitrance of biochar and of the soil N and C pools are believed to determine the mineralization of bioavailable forms of N contained in biochar (Clough et al., 2013). Therefore, we selected two soils with contrasting properties: degraded acid sandy Acrisol rich in organic matter, and alkaline C-depleted Calcisol with loamy texture, in order to study the effect of holm oak biochar on inorganic N leaching. In particular, we focused on the sorption properties of biochar and biochar-soil mixtures as well as on their effects on net N mineralization and net nitrification. Furthermore, two leaching cycles were performed in order to detect the effect of fresh biochar when applied together with mineral fertilizer (first leaching cycle) and the effect of biochar already ‘aged’ in the soil when fertilizer is applied (second leaching cycle). We hypothesized that biochar produced at temperature of 600 °C will decrease both NH_4^+-N and NO_3^--N leaching from both soil types, but that the sorption effect of biochar will be more pronounced in sandy Acrisol. Furthermore, the pH change in acid Acrisol can stimulate both N mineralization and nitrification and thus increase NO_3^--N losses or diminish the effect of biochar caused by its potential to adsorb NO_3^--N.

2. Materials and methods

2.1. Soil and biochar characterization

Two contrasting soils originated from Mediterranean climate were used in this study. Acid sandy Acrisol (FAO), characterized as clay-skeletal, kaolinitic, acid Palexerult according to Soil Taxonomy, was collected from Cañamero's raña formation in SW Spain. This soil is characterized by low pH, low content of exchangeable bases, low available phosphorus (P) content and exchange complex dominated by aluminum (Al) (Espejo, 1987; Table 1). Soil in the area is highly degraded as a consequence of long-term continuous tillage which resulted in loss of SOC and soil acidification. Second selected soil is Haplic Calcisol (FAO), corresponding to Typic Calcixerept according to Soil taxonomy, obtained from “La Chimenea” Field Station near Aranjuez (Madrid, Spain). This soil is characterized by high pH, high carbonate content and loamy texture (Table 1). Samples (0–15 cm soil depth) were transported to the laboratory, homogenized and sieved at field-moist state (< 5 mm) within three days. Part of the composite sample was air-dried and sieved to 2 mm for laboratory analysis. Soil pH and electric conductivity were determined in soil: deionized water (1:2.5 w/v) after one hour of shaking. Ammonium acetate (1 M, pH 7) method was used for cation exchange capacity (CEC) determination. Soil organic carbon content was measured by dichromate oxidation (Walkley and Black, 1934) after carbonates reaction with HCl (Calcisol). The contents of

Table 1
Selected soil and biochar properties.

Soil properties	Acrisol	Calcisol	Biochar properties	
pH	5.65	8.00	pH	10.2
Electric conductivity ($\mu S\ cm^{-1}$)	49.7	570	Electric conductivity ($\mu S\ cm^{-1}$)	940
CEC ($cmol_c\ kg^{-1}$)	2.73	8.84	TC (%)	68.2
TOC ($g\ kg^{-1}$)	25.8	9.55	TN (%)	0.67
Carbonate content (%$CaCO_3$)	n.p.	21.9	C_{ox} (%)	4.70
TN ($g\ kg^{-1}$)	1.28	0.90	Ash content (%)	3.49
WSC ($mg\ kg^{-1}$)	78.3	29.1	Carbonates content (%$CaCO_3$)	11.9
WSN ($mg\ kg^{-1}$)	19.0	49.2	WSC ($mg\ kg^{-1}$)	149
Field moisture capacity (%)	16.9	18.3	WSN ($mg\ kg^{-1}$)	93.4
Sand (%)	80.1	29.0	CEC ($cmol_c\ kg^{-1}$)	35.1
Silt (%)	6.10	42.0	NH_4-N sorption (mg NH_4-N g^{-1})	2.22
Clay (%)	13.8	29.0	NO_3-N sorption (mg NO_3-N g^{-1})	n.s.

CEC, cation Exchange capacity; TOC, total organic carbon; TN, total nitrogen; TC, total carbon; C_{ox}, dichromate oxidizable organic C; WSC, water soluble carbon; WSN, water soluble nitrogen; n.p., not present, n.s., not significant.