



Limited role of biochars in nitrogen fixation through nitrate adsorption



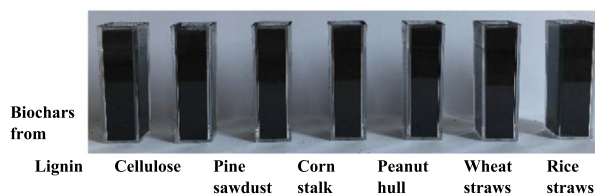
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HIGHLIGHTS

- Nitrate sorption to biochars increases with pyrolysis temperatures.
- Surface area of biochars is a key parameter controlling nitrate sorption.
- The coating of tannic acid on biochars decreases nitrate sorption.
- Biochars have comparable or lower sorption to nitrate than geosorbents.
- Biochar sorption contributes limitedly to nitrate fixation.

GRAPHICAL ABSTRACT



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ABSTRACT

Nitrate cycling is essential in sustaining soil systems. Excessive application of N-fertilizers and the associated underground water contamination have attracted a great deal of research attention. Sorption is efficient and environmentally friendly in nitrate fixation. A debate was noted in literature regarding whether biochars have potential to fix nitrate through sorption. In this study, biochars produced from different biomasses as well as biomass compositions were chosen as the adsorbents to evaluate their potential efficiencies in nitrate fixation. Increased sorption to nitrate was observed for biochars with increased pyrolysis temperature, but the increasing extent varied with biomass. The surface base functional groups and surface charges of biochars could not well explain nitrate sorption. The significant positive correlation between nitrate sorption and biochar surface areas suggested that surface area was the controlling parameter for nitrate sorption. The pre-coating of tannic acid (TA) on biochars decreased but did not completely inhibit nitrate sorption. This observation suggested that nitrate sorption on biochars may be further decreased after their interactions with natural organic matter. Nitrate sorption was compared among various adsorbents, including biochars, soil particles, clay minerals, engineered particles, as well as humic substances. Soil particles generally showed high sorption to nitrate over biochars. This result suggested that biochars investigated in this work may play a limited role in nitrate fixation through sorption after their massive application. Nitrogen fixation through nitrate adsorption on biochars should be carefully evaluated taking into consideration of biochar feedstocks and properties.

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1. Introduction

Nitrogen (N) availability controls soil ecosystems productivity. N transformation process in soils includes ammonification, nitrification,

and de-nitrification (Martens and Dick, 2003). These processes commonly generate highly mobile N forms, such as NO_3^- (nitrate), and therefore elevated N level in ground and surface water bodies (Chabani et al., 2006). The increased application of N fertilizers to soils is one of the main reasons for ground and drinking water pollution (Vitousek et al., 2009). Meanwhile, N related gases, such as N_2O , NO , and, are all reported to be greenhouse gases and contribute significantly

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to global climate change (Hansen et al., 1989). In addition, N cycling is strongly coupled with carbon cycling. For example, N inputs may stimulate organic matter decomposition (Keeler et al., 2009), which consequently increase carbon dioxide fluxes to the atmosphere. A strong negative correlation has also been reported between methane production rate and nitrate concentration level (Wang et al., 1992). Clearly, N-involved processes are closely related to greenhouse gases, and it is important to understand N behavior in soil.

Investigators have proposed various of techniques for nitrate removal, such as catalytic denitrification (Pintar and Batista, 2006), biological denitrification (Moreno et al., 2005), anion exchange (Pintar et al., 2001) and electro dialysis (Elmidaoui et al., 2001). With the recent and increasing attention on biochars (Lehmann et al., 2006), it has been proposed that biochars could be an effective and cheap material for N immobilization through direct sorption of nitrates (Singh et al., 2010). For example, sugarcane bagasse may adsorb nitrate through the interaction with base functional groups (Kameyama et al., 2012). Wood biochars decreased nitrate leaching due to their anion retention capacity (Dempster et al., 2012). When three kinds of biomass (corn stover, ponderosa pine wood chips and switchgrass) were pyrolysed to form biochars, the surface area and net surface charge showed significant effects on nitrate sorption (Chintala et al., 2013).

This sorption-based concept has been successfully used in reducing nitrate leaching in soils. Biochars (made by wheat straw at 600 °C) amended red soil in China showed reduced nitrate leaching (Jin et al., 2016). A commercial charcoal derived from rice husk could adsorb nitrate of 2.1 mg/g, which consequently reduced nitrate leaching by 23% in soil (Pratiwi et al., 2016). Nitrate retention by wood chips-derived biochars was also explained by water-ion hydrogen-bonding in biochar porous surface (Kammann et al., 2015). In a biochar-amended topsoil, nitrate retention increased even after the long term harvest (1 year and 2 year) when mostly nitrate was supposed to have been taken up by the crop or leached out of the topsoil (Haider et al., 2016). A wood-

based biochar was applied to a macadamia orchard soil. The soil showed significantly increase of nitrate retention after 12-month biochar application (Bai et al., 2015). The authors emphasized abiotic factors with the aid of stable isotope experiments. Prendergast-Miller et al. (2011) observed accumulation of nitrate in a biochar amended rhizosphere soil. The authors suggested that nitrate may be localized in the pore spaces of biochars in the rhizosphere.

However, biochar is known to be negatively charged and the mechanism for such sorption is unclear. Some studies show limited or no sorption of nitrate. Hollister et al. (2013) observed no adsorption of nitrate to biochar derived from corn or oak because of insufficient anion exchange capacity. Yao et al. (2012) found that nine of thirteen biochars had little nitrate adsorption capacity due to their net negative surface charge. The about-mentioned limited sorption may be one of the reasons that the application of biochar in Canterbury, New Zealand under dairy winter forage grazing conditions showed no significantly reduction of nitrate leaching (Hill et al., 2014). Previous studies also showed that biochar surfaces may be oxidized to generate acidic functional groups, which may further decrease their sorption to nitrate (Cheng et al., 2008; Lin et al., 2012).

With an increasing body of studies, the mechanisms of nitrate immobilization on biochar and the influence of natural environment are still under debate. Knowing that the practical technique is largely dependent on the understanding of N sorption mechanisms, extended researches are needed to reveal the sorption mechanisms of various forms of N in biochars.

This study was thus designed to investigate the properties of biochars produced from various feedstocks in relation to nitrate fixation. Our hypothesis is that biochar sorption to nitrate is low and thus contributes limitedly to N fixation. Batch sorption experiments were conducted in this study. Biochars were systematically characterized using BET adsorption technique, Fourier transform infrared (FTIR) spectra and zeta potential. Nitrate sorption on natural geosorbents was also collected for comparison.

Table 1
BET surface area, pore size distribution and elemental composition of biochars.

Sample ^a	$a_{s,BET}$ [m ² g ⁻¹]	Total pore volume ($p/p_0 = 0.990$) [cm ³ g ⁻¹]	Average pore diameter [nm]	Elemental composition (%)				
				N	C	H	S	O
L-RT	2.01	0.00624	12.4	0.118	43.7	4.50	4.16	31.1
L-200	1.84	0.00578	12.5	0.116	54.4	4.89	4.55	32.0
L-350	0.710	0.00292	16.4	0.158	62.6	3.27	4.14	22.5
L-500	208	0.0855	1.64	0.259	72.1	2.03	5.02	14.9
L-700	201	0.0976	1.94	0.419	63.8	0.912	5.73	19.8
L-500-TA	3.65	0.00595	6.52	0.385	85.8	2.45	1.79	9.30
C-RT	0.822	0.00614	29.9	0.060	41.7	6.17	0	48.3
C-200	0.830	0.00560	27.0	0.033	44.0	6.54	0.084	49.2
C-350	0.637	0.00341	21.4	0.075	81.2	4.26	0.110	15.9
C-500	412	0.183	1.78	0.113	88.3	3.12	0.099	7.34
C-700	881	0.380	1.72	0.124	85.3	2.97	0.083	8.92
C-500-TA	6.09	0.00538	3.53	0.230	89.5	3.10	0.143	9.63
PS-RT	–	–	–	0.087	50.9	5.92	0.173	42.4
PS-200	0.493	0.00169	13.7	0.079	51.4	6.28	0.076	40.7
PS-350	2.14	0.00528	2.83	0.268	73.7	4.48	0.127	21.5
PS-500	19.8	0.0167	3.73	0.238	85.2	2.86	0.068	12.7
PS-700	566	0.302	2.13	0.250	79.7	1.55	0.133	11.0
PS-500-TA	3.37	0.00631	7.48	0.390	82.3	2.94	0.167	13.4
CS-RT	–	–	–	0.650	45.5	6.08	0.244	42.0
CS-500	71.4	0.452	2.53	0.934	71.9	2.43	0.218	13.1
CS-500-TA	18.1	0.0284	6.28	1.22	75.8	2.47	0.0871	14.0
WS-RT	–	–	–	0.528	44.3	5.88	0.172	42.2
WS-500	113	0.0878	2.77	0.749	67.3	2.36	0	13.6
WS-500-TA	58.3	0.0429	2.94	0.722	68.5	2.46	0.203	12.6
RS-RT	1.20	0.00507	16.9	0.898	39.7	5.58	0.077	39.2
RS-500	111	0.0622	2.23	0.932	53.1	2.11	0.778	13.0
PH-RT	0.350	0.00311	35.6	1.66	44.6	5.89	0.099	39.0
PH-500	227	0.0953	1.68	1.25	75.5	2.46	0.122	12.8

^a L for lignin, C for cellulose, PS for pine sawdust, CS for corn straw, WS for wheat straw, PH for peanut hull, RS for rice straw. The numbers behind the dash are the pyrolysis temperatures. TA means the biochars were coated with tannic acid. The symbol of “–” indicates values below detection limit.

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