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Biochar for the mitigation of nitrate leaching from soil amended with biosolids

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ABSTRACT

Countries with sewage treatment plants produce on average 27 kg of dried biosolids/person/yr. Concerns about nitrate leaching limit the rate at which biosolids are added to soil. We sought to determine whether biochar, a form of charcoal that is added to soil, could reduce nitrate leaching from biosolids amended soil. We set up 24 (0.5 m × 0.75 m) lysimeters, filled with two soil types (Templeton Silt Loam and Ashley Dene silt loam) and amended with combinations of biochar (102 t/ha equivalent) and biosolids (600 and 1200 kg N/ha equivalent). Pasture and leachates were sampled over 5 months. Nitrate leaching from biochar plus biosolids amended soils were reduced to levels at or below the control treatments. Pasture N concentrations were similarly affected by biochar addition. Future research should focus on unravelling the mechanism responsible for the change in the nitrogen cycle in soils amended with biosolids and biochar.

1. Introduction

Countries with sewage treatment plants produce some 27 kg of dry biosolids per person per year, with global output exceeding 10 M t/yr (Bradley, 2008). Biosolids disposal is a global environmental issue, with many developed countries incinerating their biosolids or disposing of them in landfills. Land application of biosolids can improve soil fertility and reduce the need for mineral fertilisers (Obi and Ebo, 1995). Depending on their provenance and level of treatment, biosolids may contain high concentrations of macro and micro-nutrients, organic compounds, heavy metals, endocrine disrupting compounds, pesticides, herbicides, surfactants, and pathogenic helminths, bacteria, viruses and fungi (Krogmann et al., 1999; Singh and Agrawal, 2008). Therefore, depending on their quality, biosolids addition to soil can result in contaminant accumulation (Stoven and Schnug, 2009), human exposure to pathogens, and leaching of plant nutrients, particularly nitrates, into groundwater (Agopsowicz et al., 2008). Stricter regulations and improved treatment technology have resulted in reduced pathogen burdens and decreased concentrations of heavy metals and organic contaminants (Chaney, 1990), leaving N loading as the rate limiting factor of the addition of high-quality biosolids in some countries (Gibbs, 2003). Nitrate leaching is also an issue, when biosolids (either high or low quality) are used to rebuild degraded soils, since higher application rates are required (Kowaljow et al., 2010).

Biochar, a form of charcoal that is added to soil (Lehmann et al., 2003), may be a potential solution. Biochar addition to soil is currently generating much interest due to its perceived potential to offset human-induced global climate change emissions (Clough and Condron, 2010; Gaunt and Lehmann, 2008; Laird, 2008; Lehmann et al., 2006; Rondon et al., 2005). However, there may be additional agronomic benefits. Biochar improves soil porosity (Steiner et al., 2007; Yanai et al., 2007), surface area (Laird et al., 2010a, 2010b), and decreases tensile strength thus improving root penetration (Chan et al., 2007). Chemically, biochar may increase cation and anion exchange capacity (CEC & AEC) (Liang et al., 2006; Singh et al., 2010), and soil pH (Cheng et al., 2006). Biochar additions in combination with nutrient applications containing available N in the form of bovine urine, swine manure and green-waste compost has interfered with the N cycle (Beesley et al., 2010; Clough et al., 2010; Laird et al., 2010b). Biochar derived from pecan shells has been demonstrated to reduce nitrate leaching from soil over 25 and 67 days (Novak et al., 2010). Deenik et al. (2010) proposed that volatile matter in biochar may stimulate soil microbial activity, which consumes nitrogen, rendering it unavailable for plant uptake. However, other authors (Beesley and Dickinson, 2011; Beesley et al., 2010) have shown that biochar may contain agents such as polycyclic aromatic hydrocarbons, that can be detrimental to microbial growth.

A potential source material for biochar is the Monterey Pine (*Pinus radiata* D. Don), which has been introduced as a timber tree in vast areas of New Zealand (where it is the most common tree), Australia, Chile, South Western Europe and South Africa (Earle, 2010). Waste wood from timber operations is ether burned or stored in large wood-waste piles than can leach tannins into local waterways (Robinson et al., 2007). We do not propound the use of treated timber for biochar production as this could result soils becoming contaminated with Cu,

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Cr, As or B. Producing bicohar from the Monterey Pine rather than biosolids themselves has the advantage that the wood waste has a lower moisture content, thus producing more energy and a higher yield than moist biosolids. The C: N ratio of Monterey Pine wood is 400 (Robinson et al., 2003), some 20 times higher than biosolids. Therefore, the Monterey Pine biochar will not add significant amounts of nitrogen to the biosolids-amended soil, which may already have a high nitrogen load. Taghizadeh-Toosi et al. (2011) demonstrated that biochar made from Monterey pine interfered with the nitrogen cycle and reduced nitrous oxide emissions from bovine urine patches.

Soils under Monterey Pine forestry can become depleted in nutrients and harvesting may remove topsoil and organic matter (Merino and Edeso, 1999). Therefore, such soils are ideal candidates for biosolids and biochar application, which would increase both soil carbon and plant nutrients. The costs of transporting the biochar would be minimal since the wood-waste is produced on site.

We aimed to determine the effect of biochar made from Monterey Pine on the leaching and plant uptake of N from biosolids amended soils, using two biosolids application rates (600 kg N/ha and 1200 kg N/ha) and two soil types.

2. Materials and methods

2.1. Lysimeter setup

Fifteen undisturbed soil monolith lysimeters were collected from a pasture soil, the Templeton Silt Loam (TSL), on the Lincoln University Dairy Farm (43°38′40.89″ S 172°26′24.01″ E) and nine were collected from a former Monterey Pine forestry plantation soil, the Ashley Dene Silt Loam (ASL), on the Ashley Dene Sheep Farm (43°39′05.82″ S 172°19′41.47″ E). Table 1 gives the properties for each soil. Each lysimeter was 0.5 m in diameter and 0.7 m deep. The design of the lysimeter casings and the method of soil sampling are described in detail by Cameron et al. (1992). The lysimeters were installed at the Field Service Centre, Lincoln University (43°38′53.48″S 172°28′07.58″ E) in a block design beside a trench used for drainage collection.

Biosolids were obtained from the Kaikoura Regional treatment works, New Zealand. Some 160 kg of biosolids were homogenised using a concrete mixer and passed through a 20 mm sieve. The moisture content of the fresh biosolids was 53%. Table 1 gives the properties of the biosolids. The biochar was manufactured from *Pinus radiata* D. Don as described in Clough et al. (2010) and Taghizadeh-Toosi et al. (2011). The biochar was crushed to give particles with a maximum diameter of 10 mm. Table 1 gives the properties of the biochar.

Table 1
Soil chemical properties for the Templeton and Ashley Dene silt loams, the biosolids and biochar. Values in brackets represent the standard error of the mean (n = 3 unless otherwise indicated).

	Ashley Dene	Templeton	Biosolids	Biochar
pH	5.6	5.6	4.1	7.8
CEC ($cmol_{(+)}$/kg)	11.9 (0.2)	12.4 (0.5)	n.d.	8.0[‡*]
C (%)	2.4 (0.1)	2.0 (0.1)	28.0 (0.2)	70.6[‡]
N (%)	0.28 (0.01)	0.18 (0.01)	2.7 (0.03)	0.2[‡]
P (mg/kg)	784 (39)	518 (25)	4683 (2)	412 (2)
S (mg/kg)	266 (18)	193 (15)	6972 (43)	288 (12)
Ca (mg/kg)	3037 (242)	3005 (101)	9818 (176)	7758 (160)
Mg (mg/kg)	802 (2)	855 (11)	2204 (17)	605 (11)
K (mg/kg)	1438 (152)	1401 (119)	4330 (67)	1713 (17)
Na (mg/kg)	113 (10)	136 (4)	428 (3)	10000 (29)
Cd (mg/kg)	0.2 (0.0)	0.4 (0.1)	2.8 (0.0)	0.1 (0.01)
Cr (mg/kg)	8.5 (0.6)	11.6 (0.4)	32 (1.4)	2.8 (0.6)
Cu (mg/kg)	5.7 (0.3)	4.5 (0.1)	561(33)	14 (5)
Pb (mg/kg)	7.7 (0.0)	12.0 (0.1)	96 (3)	1.0 (0.2)
Zn (mg/kg)	35 (1)	43 (1)	878 (13)	16 (1.3)

[‡]Single analysis on homogenised material.
[*]Taghizadeh-Toosi et al. (2011).

There were eight treatments, each replicated three times. For both the TSL and ASL lysimeters, there was a control (no bisolids, no biochar), fresh biosolids applied at a rate equivalent to 600 kg N per hectare (0.9 kg biosolids, no biochar), and biosolids with biochar at a rate equivalent to 100 t per hectare (0.9 kg biosolids, 2 kg biochar). The TSL lysimeters had two additional treatments of biosolids applied a rate equivalent to 1200 kg N per hectare (1.8 kg biosolids, no biochar) and biosolids with biochar (1.8 kg biosolids, 2 kg biochar). Our rate of biochar addition was at the top end of rates reported by other authors (Chan et al., 2007). The rational for this was that this biochar – biosolids mixture would be added to rebuild low-fertility soils, where increasing soil carbon and soil nutrients is paramount.

The top 0.1 m of the soil profile was removed from all lysimeters. Treatments were applied by mixing the soil with the biosolids & biochar in a concrete mixer and refilling the top 0.1 m of the lysimeter. The controls were handled in an identical manner, without any treatment. Before refilling, a soil sample was taken for analyses. Refilling occurred on the 5 May 2010. Immediately after the application of each treatment, pasture ryegrass (*Lolium perenne* Bronsyn) was broadcast over each lysimeter to give a density of ca. 200 seeds per dm^2. A small amount of soil from each treatment was applied over the top of the seed and lightly pressed down by hand.

2.2. Climatic conditions and irrigation

Irrigation was applied during the initial two weeks to initiate drainage. Thereafter the only influent was natural rainfall. Total water inputs during the trial period were equivalent to 574 mm over five months (Fig. 1). This comprised of 547 mm of natural rainfall and 27 mm of irrigation.

2.3. Sample collection and chemical analyses

Drainage was collected fortnightly from the base of each lysimeter. The volume of drainage was measured and recorded during each sampling. A 100 mL subsample was taken for analysis. Pasture was harvested on the 31st of August and the 24th of September 2010. The timing of harvest occurred upon the emergence and full development of the 3rd leaf. This corresponds to maximal pasture quality, with no leaf death and fully developed leaves. The pasture was harvested to a height of 20 mm above the soil level.

Each drainage sample was vacuum filtered with a water aspiration apparatus through 0.45 μm cellulose acetate filter membrane. Nitrates and nitrites were measured using ion chromatography. Organic C was analysed using a Shimadzu TOC 5000A organic carbon analyser.

Soil and pasture samples were dried at 105 °C until a constant weight was obtained. Large soil aggregates were broken up using a mortar and pestle and the soil passed through a 2 mm Nylon sieve.

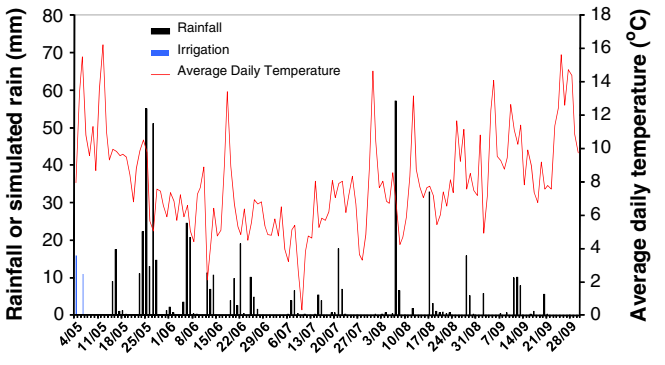

Fig. 1. Rainfall, irrigation and average daily temperature during the five-month lysimeter trial.

Soil pH was measured at a water: soil ratio of 2.5:1 with deionised water using a Metler Toledo pH meter.

Dried pastures were ground and stored in an airtight vial. Soil and Pasture C and N concentrations were measured using an Elementar Vario MAX CN analyser. Pseudo-total elemental analysis was carried out using microwave digestion in 8 ml of Aristar nitric acid (±69%), filtered using Whatman 52 filter paper (pore site 7 μm), and diluted with milliQ water to a volume of 25 ml, followed by analysis by inductively coupled plasma optical emission spectrometry (ICP-OES Varian 720 ES). Wageningen reference soil (ISE 921) and plant (IPE 100) material were analysed for quality assurance (Houba et al., 1998). Concentrations were within 9% of the certified values.

Data was analysed using Minitab® 16. Data sets were analysed using ANOVA with Fisher's Lest-Significant-Difference post-hoc test to compare means. The level of significance was 0.05.

3. Results and discussion

3.1. Pasture growth and nitrogen uptake

Pasture established full cover on all the lysimeters (Table 2), with biomass productions of the ASL and TSL controls being equivalent to 1.4 tonnes of dry matter per hectare. There were significant differences in pasture growth between treatments (Table 2). The addition of biosolids, without biochar, resulted in a significant biomass increase in both the ASL and TSL treatments. This result is consistent with the fertilising effect of the biosolids, which contain high concentrations of N and P. In the ASL, there were no significant differences in the N concentration (Table 2) of the pasture. However, in the TSL, the 1200 N treatment resulted in a significant increase in the pasture N concentration.

The addition of biochar lessened or negated the biomass increases resulting from biosolids addition. In the 600 N treatment with biochar, the biomass was not significantly different to the control, while in the TSL, the addition of biosolids at 1200 kg/ha with biochar produced a significant increase that was less than the biosolids treatment alone. The effect of the biochar in lessening the biomass increase may be attributed to either a toxic factor in the biochar that reduced growth, or that the biochar absorbed some of the nutrients provided by the biosolids, thereby lessening their effect. We cannot definitively rule out the possibility that the biochar treatments inhibited growth through some toxic factor because we did not have a treatment where biochar was added in the absence of biosolids.

3.2. Drainage and nitrogen leaching

The volume of drainage was equivalent to 426 mm and 412 mm for the TSL and ASL controls respectively, indicating that the pasture evapotranspiration was 148 mm and 162 mm respectively. Despite the significant differences in biomass (Table 2), none of the treatments had significantly different drainage volumes compared to the control. This is unsurprising given the relatively low evaporative demand during the winter months and that the grass had established a complete cover on all the lysimeters, providing similar surface areas for evapotranspiration.

The N leached from the lysimeters was predominantly in the form of nitrate, with concentrations in the drainage ranging from 9 to 35 mg/L. Concentrations of nitrite and ammonium were <0.3 mg/L. Dissolved organic carbon concentrations in the leachate were <35 mg/L. The concentration of dissolved organic nitrogen (not determined) depends on a number of factors including land use and vegetation (Bolan et al., 2011). Using a C:N ratio in dissolved organic matter of 25 (Wu et al., 2010) the concentrations of dissolved organic N in our study are likely to be <1.5 mg/L.

Fig. 2(A and B) shows the nitrate leaching from the ASL and TSL lysimeters as a function of the cumulative leachate volume. For all lysimeters, the cumulative nitrate leaching vs cumulative drainage produced sinusoidal curves, with points of inflection around 10 L and 50 L of cumulative drainage. This is consistent with a breakthrough of a pulse of nitrate, resulting from the application of disturbed soil and treatment mixtures. There were significant differences between the treatments. Nitrogen losses from the ASL treatments were significantly higher than the corresponding treatments in the TSL treatments. This is probably due to the higher N concentration in the ASL soil (Table 1). The addition of biosolids resulted in a significant increase in nitrate leaching in all treatments, except the TSL 600 N. This is consistent with the high N concentration in the biosolids. In the field situation, this could lead to groundwater contamination, hence the environmental legislation limiting the rate of biosolids application to soil.

The addition of biosolids and biochar together resulted in nitrate leaching that was significantly lower than the biosolids alone treatment, and the 600 N char treatment leached significantly less nitrate than the control. This indicates that by including biochar in a biosolids soil amendment can mitigate nitrate leaching from over the short term.

Table 2
The above-ground biomass production of the pasture and its average N concentration during the lysimeter (0.2 m^2) experiment. Values in brackets are the standard error of the mean. Values in a column with the same letter are not significantly different.

Treatment	Dry biomass (g)	Average N concentration (%)
ASL control	24.7 (3.2)^{d}	2.44 (0.08)^{d}
ASL 600 N	32.5 (1.5)^{bc}	2.52 (0.10)^{cd}
ASL 600 N char	21.5 (0.7)^{d}	2.57 (0.07)^{bcd}
TSL control	27.1 (0.6)^{cd}	2.55 (0.03)^{cd}
TSL 600 N	36.1 (0.6)^{b}	2.86 (0.03)^{abc}
TSL 600 N char	27.3 (1.8)^{cd}	3.04 (0.26)^{c}
TSL 1200 N	42.3 (3.0)^{a}	2.95 (0.07)^{ab}
TSL1200N char	35.2 (2.2)^{b}	2.89 (0.09)^{ab}

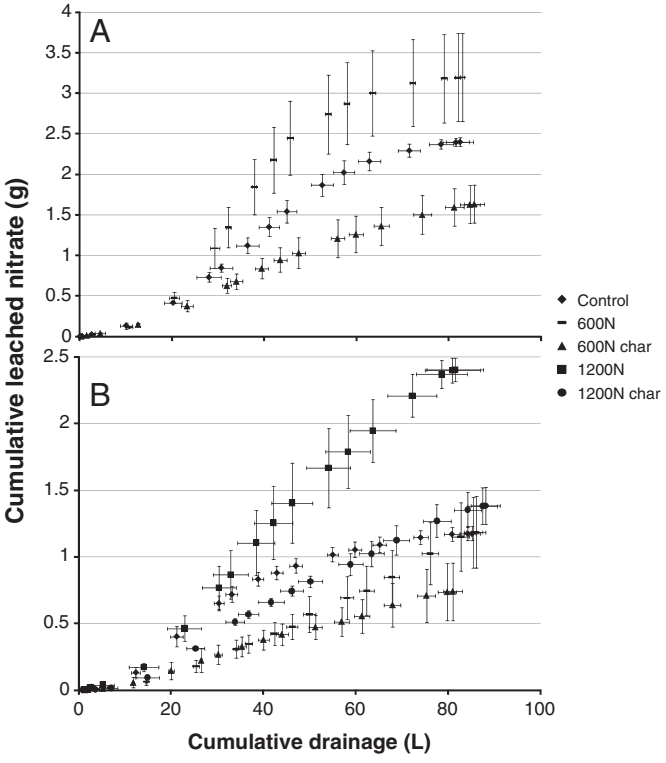

Fig. 2. Cumulative mass of leached nitrate (g) vs cumulative drainage volume (L) from lysimeters (0.2 m^2) for the Ashley Dene silt loam (A) and the Templeton silt loam (B). The error bars represent the standard error of the mean.

Table 3
Mass (g) of N present initially in lysimeters (0.2 m^2), N removed via plant uptake and nitrate removed via leaching. Values in brackets are the standard error of the mean. Values in a column with the same letter are not significantly different.

	Nitrogen present initially		Nitrogen removed		*% removed*
	Soil (top 100 mm)	Added in biosolids	Plant uptake	Leached	
ASL control	42.0	0.0	0.6 (0.1)^{e}	2.4 (0.1)^{b}	7.1
ASL 600 N	42.0	11.8	0.8 (0.1)^{cd}	3.2 (0.5)^{a}	7.4
ASL 600 N char	42.0	11.8	0.6 (0.0)^{e}	1.6 (0.2)^{c}	4.1
TSL control	27.0	0.0	0.7 (0.0)^{de}	1.2 (0.1)^{cd}	4.4
TSL 600 N	27.0	11.8	1.0 (0.0)^{ab}	1.2 (0.1)^{cd}	5.7
TSL 600 N char	27.0	11.8	0.8 (0.1)^{bcd}	0.7 (0.2)^{d}	3.9
TSL 1200 N	27.0	23.6	1.2 (0.1)^{a}	2.4 (0.1)^{b}	7.1
TSL 1200 N char	27.0	23.6	1.0 (0.1)^{bc}	1.4 (0.1)^{cd}	4.7

3.3. Nitrogen losses from the system

Table 3 shows the mass of N initially present in each lysimeter as well as N removed from the system via plant uptake and via nitrate leaching. The amount of nitrogen lost from the system via leaching will be slightly higher because this figure does not include other nitrogen species, which as discussed above, are insignificant compared to nitrate. Significantly more N was removed via plant uptake and leaching in the biosolids treatments compared to the control, while N removed from the biosolids + biochar treatments was not significantly different from, or significantly less than the control. The total N removed as a percentage of that initially present in the top 100 mm ranged from 3.9% (TSL 600 char) to 7.4% (ASL 600). On a per-hectare basis, the N leached during the experiment ranged from 36 kg/ha (TSL600 char) to 163 kg/ha (ASL600). The values from the ASL and TSL controls, 122 and 61 kg/ha are similar to those reported by Cameron and Wild (1984) for a ploughed pastureland (up to 200 kg/ha), but significantly higher than non-ploughed pastureland (Di and Cameron, 2002). Since we homogenised the top 100 mm of the soil, we expected our lysimeters to behave similarly to a ploughed field. That there was no significant difference in the N leached between the TSL control and TSL 600 treatment may be due to the increased N uptake by pasture on the latter (Table 3).

We did not quantify N losses through volatilisation, which may have occurred as NH_3, N_2, or N_2O. Given the low pH of the soil (5.7), we would expect NH_3 losses to be negligible. Losses of N_2 are environmentally benign, while N_2O is a potent greenhouse gas. Previous studies have shown that biochar decreases N_2O emissions from urine patches in pasture (Taghizadeh-Toosi et al., 2011) and soil amended with poultry manure (Singh et al., 2010).

Biochar could mitigate nitrate leaching from biosolids by inhibiting the mineralisation of organic N to ammonia and thence to nitrate or by sorbing ammonium or nitrate, thus rendering it less available for leaching and plant uptake. If sorbtion was occurring, the mechanisms for this are unclear. The CEC of the biochar (Table 1) was less than that either the ASL or TSL soils, indicating that retention of ammonium via this mechanism will not be significant. The AEC of the biochar was just 4.0 $cmol_{(+)}$/kg (Taghizadeh-Toosi et al., 2011), ruling out significant electrostatic binding of nitrate. Potentially, soil solution containing ammonium and nitrate could have been incorporated into pores on the surface of the biochar, which had a porosity of 0.64 (Taghizadeh-Toosi et al., 2011). However, such pores would have rapidly become saturated (Fig. 2) shows no significant differences in leachate volume and further mineralised N would have leached. Alternatively, the biochar may have inhibited the growth of soil flora that normally mineralises and nitrifies N. This could have occurred through some toxic agent on the surface of the biochar (Kim et al., 2003), or by providing refugia (Warnock et al., 2007) for competing microorganisms or denitrifying bacteria. It is interesting to note that the needles of Monterey Pine contain toxic polyphenols and flavonoids (Adams et al., 1992). These compounds may have also been present in the wood from which the biochar was manufactured.

4. Conclusions

The incorporation of biochar into biosolids-amended soil mitigates nitrate leaching over the short term. This delay should be beneficial both to the environment, which receives lower nitrate loadings, and to plants, which have N held in the rootzone for longer periods. Biosolid/biochar mixtures could be added to soils at a much higher rate than biosolids alone, thereby increasing the efficacy of using biosolids to rebuild degraded soils, where organic matter is limiting. There are several unknowns resulting from this study that warrant further research. The mechanisms for the inhibition of nitrate leaching are unclear. Elucidating these may enable optimisation of bisolids: biochar ratio and allow the manufacture of biochars with the best properties for the reduction of nitrate leaching.

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