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Optimum rates of surface-applied coal char decreased soil ammonia volatilization loss

Dinesh Panday[1] | **Maysoon M. Mikha**[2] | **Harold P. Collins**[3] | **Virginia L. Jin**[4] | **Michael Kaiser**[1] | **Jennifer Cooper**[1] | **Arindam Malakar**[5] | **Bijesh Maharjan**[1]

[1]Dep. of Agronomy and Horticulture, Univ. of Nebraska-Lincoln, Lincoln, NE 68588, USA

[2]USDA-ARS, Central Great Plains Research Station, Akron, CO 80720, USA

[3]USDA-ARS, Grassland Soil and Water Research Laboratory, Temple, TX 76502, USA

[4]USDA-ARS, Agroecosystem Management Research Unit, Lincoln, NE 68583, USA

[5]Nebraska Water Center, Univ. of Nebraska-Lincoln, Lincoln, NE 68588, USA

Correspondence

Bijesh Maharjan, Dep. of Agronomy and Horticulture, Univ. of Nebraska-Lincoln, Lincoln, NE 68588, USA.

Email: bmaharjan@unl.edu

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Abstract

Fertilizer N losses from agricultural systems have economic and environmental implications. Soil amendment with high C materials, such as coal char, may mitigate N losses. Char, a coal combustion residue, obtained from a sugar factory in Scottsbluff, NE, contained 29% C by weight. A 30-d laboratory study was conducted to evaluate the effects of char addition on N losses via nitrous oxide (N_2O) emission, ammonia (NH_3) volatilization, and nitrate (NO_3–N) leaching from fertilized loam and sandy loam soils. Char was applied at five different rates (0, 6.7, 10.1, 13.4, and 26.8 Mg C ha^{-1}; char measured in C equivalent) to soils fertilized with urea ammonium nitrate (UAN) at 200 kg N ha^{-1}. In addition, there were two negative-UAN control treatments: no char (no UAN) and char at 26.8 Mg C ha^{-1} (no UAN). Treatment applied at 6.7 and 10.1 Mg C ha^{-1} in fertilized sandy loam reduced NH_3 volatilization by 26–37% and at 6.7, 10.1, and 13.4 Mg C ha^{-1} in fertilized loam soils by 24% compared with no char application. Nitrous oxide emissions and NO_3–N leaching losses were greater in fertilized compared with unfertilized soil, but there was no effect of char amendment on these losses. Because NO_3–N leaching loss was greater in sandy loam than in loam, soil residual N was twofold higher in loam than in sandy loam. This study suggests that adding coal char at optimal rates may reduce agricultural reactive N to the atmosphere by decreasing NH_3 volatilization from fertilized soils.

1 | INTRODUCTION

Fertilizer nitrogen (N) use increased globally at an annual rate of 1.4% from 2014 to 2018 (IFASTAT, 2019). Generally, crop N uptake efficiency is <50% of applied N, which leaves a significant amount of N in soil prone to loss via NH_3 volatilization, NO_3–N leaching, and/or denitrification as N_2O emissions (Fageria & Baligar, 2005; Robertson et al., 2013). Nitrogen losses from agricultural systems can be a major limitation for crop production and environmental sustainability.

Abbreviations: C0N0, no char or urea ammonium nitrate; C0N1, no char and urea ammonium nitrate; C1N1, char rate at 6.7 Mg C ha^{-1} and urea ammonium nitrate; C2N1, char rate at 10.1 Mg C ha^{-1} and urea ammonium nitrate; C3N1, char rate at 13.4 Mg C ha^{-1} and urea ammonium nitrate; C4N1, char rate at 26.8 Mg C ha^{-1} and urea ammonium nitrate; C4N0, char rate at 26.8 Mg C ha^{-1} and no urea ammonium nitrate; CCR, coal combustion residue; CEC, cation exchange capacity; CV, coefficient of variance; FNR, fertilizer N recovery; OM, organic matter; UAN, urea ammonium nitrate.

Numerous management technologies have been proposed to mitigate N losses from agricultural systems, including the proper management of soil C because of its effects on soil properties and processes, including N cycling (Dil, Oelbermann, & Xue, 2014; Ding et al., 2010). Carbon management practices that include amendments with high C content, such as biochar, can boost soil fertility and quality by raising pH and by improving water holding capacity, cation exchange capacity (CEC), and nutrient retention (Bridgwater, 2003; Filiberto & Gaunt, 2013; Singh et al., 2014).

Coal combustion residues (CCRs), such as fly ash, bottom ash, and flue gas desulfurization gypsum, have been used as soil amendments to improve soil health and crop performance (Basu, Pande, Bhadoria, & Mahapatra, 2009; Panday, Ferguson, & Maharjan, 2018; Shaheen, Hooda, & Tsadilas, 2014). However, depending on the composition and nature of CCR, they can enhance mineralization of organic soil N and N losses (Siddaramappa, McCarty, Wright, & Codling, 1994). The CCRs in electric power generating stations obtained from the near-complete combustion of coal during energy production contain very little C. In contrast, coal char (henceforth "char") resulting from inefficient coal burning can contain up to 29% C by dry weight as well as other essential plant mineral nutrients.

Char stands midway between coal ash and biochar with respect to C content. Biochar and other hydrocarbons are typically produced from pyrolysis of biomass in the presence of little or no oxygen at a range of temperatures and can contain up to 70% of initial biomass C (Atkinson, Fitzgerald, & Hipps, 2010; Lehmann, Gaunt, & Rondon, 2006). Biochar can reduce NH_3 volatilization loss (Steiner, Das, Melear, & Lakly, 2010) and NO_3–N leaching loss (Hagemann, Kammann, Schmidt, Kappler, & Behrens, 2017). However, the beneficial effect of biochar in reducing environmental N losses from fertilized soil is not consistent and depends on sources and production conditions (Ding et al., 2016). Char, which is different from regular CCRs and biochar but has a considerable amount of C, warrants exploration for its potential use in agricultural soil.

The objectives of this study were to evaluate the effects of char on soil N losses in the form of NH_3 volatilization, N_2O emissions, and NO_3–N leaching from fertilized loam and sandy loam soils. We hypothesized (a) that adding char would reduce N losses from fertilized soil by improving the retention of applied N and (b) that char effectiveness on retaining N would differ by soil type.

Core Ideas

- High C content coal char may reduce environmental N loss from fertilized soil.
- There are implications of using different methods in estimating fertilizer N recovery.
- Must evaluate industrial by-products in agriculture for potential accumulation of trace metals.

2 | MATERIALS AND METHODS

The char used in this study was a CCR from a sugar factory in Scottsbluff, NE, and contained 29.3% C and some nutrients (Supplemental Table S1). It also contained heavy metals (As, Cd, Cr, Pb, Hg, and Se), but their concentrations were below the USEPA's ceiling limits for heavy metal soil contamination or phytotoxicity in soil (Cameron, 1992). Char was sieved through a 2-mm sieve. The physical characteristics of char were determined by X-ray diffraction using a PANalytical Empyrean Diffractometer (Malvern Panalytical Ltd.) at the Nebraska Center for Materials and Nanoscience (Supplemental Figure S2). Brunauer-Emmett-Teller surface area of char was analyzed with an ASAP 2460 Surface Area and Porosity Analyzer (Micromeritics Instrument Corporation) at the Nebraska Center for Materials and Nanoscience (Supplemental Table S3).

Two soils were used to evaluate the effects of char on N losses from fertilized soil at the Panhandle Research and Extension Center, University of Nebraska-Lincoln in Scottsbluff, NE. One soil was a Tripp fine sandy loam (coarse-silty, mixed, superactive, mesic Aridic Haplustolls, 0–3% slope) with pH 7.7; 13 g kg^{-1} organic matter (OM); and 60, 28, and 12% of sand, silt, and clay contents, respectively. This soil was collected from the Panhandle Research and Extension Center. The other soil was a Duroc loam (fine-silty, mixed, mesic Pachic Haplustolls, 0–1% slope) with pH 7.2; 18 g kg^{-1} OM; and 40, 33, and 27% of sand, silt, and clay, respectively. This soil was collected from farmland near the University of Nebraska-Lincoln High Plains Agricultural Laboratory in Cheyenne, NE. Both soils were collected at depths of 0–20 cm in the spring of 2018. Residual inorganic N rates, extracted with 2 M KCl, in loam and sandy loam soils were 5.2 and 3.7 mg kg^{-1}, respectively.

Collected soils were air-dried and sieved through a 2-mm mesh. Soils were brought to 10% gravimetric water content (GWC) by applying water and mixing thoroughly, which corresponded to 70 and 50% of field capacity of sandy loam and loam, respectively. Soils were packed in 5-cm-diameter clear acrylic columns (Supplemental Figure S4) to a height of 24 cm with a targeted bulk density of 1400 kg m^{-3} (Peng et al., 2015). A porous ceramic plate (0.1 MPa strength) was inserted in the bottom of the column and topped with Whatman no. 42 filter paper to prevent soil from clogging the ceramic plate. Soil columns had lid systems at either end. A vacuum port located on the bottom lid allowed suction to be applied during the collection of leachate. The top lid has two parts (lower and upper). The lower lid part is an elongated connector (height, 5 cm) threaded onto the main column and the upper lid, which was used to install the NH_3 acid trap. The upper lid part (height, 5 cm) terminates the column with a closed end fitted with a septum port for N_2O sampling from the headspace above the soil.

Char (measured in C equivalent) and UAN were applied to each soil column and mixed in the top 6-cm soil layer. There were seven treatments, each with four replications: (a) C0N0, no char or UAN; (b) C0N1, no char and UAN; (c) C1N1, char rate at 6.7 Mg C ha^{-1} and UAN; (d) C2N1, char rate at 10.1 Mg C ha^{-1} and UAN; (e) C3N1, char rate at 13.4 Mg C ha^{-1} and UAN; (f) C4N1, char rate at 26.8 Mg C ha^{-1} and UAN; and (g) C4N0, char rate at 26.8 Mg C ha^{-1} and no UAN. All treatments that were fertilized (CxN1) received 39.5×10^{-3} g UAN-N that was equivalent to 200 kg N ha^{-1}.

After soil columns were prepared, water was periodically added to simulate rainfall (100.8 mm in total) in May 2017 in Scottsbluff, NE (Supplemental Figure S5). Water was added slowly on the surface of soil using a syringe to prevent ponding on the surface. Columns were kept on the laboratory benchtop at constant room temperature (25°C) throughout the 30-d experimental period.

2.1 | Sample collection

Ammonia volatilization was measured using an acid trap method (McGinn & Janzen, 1998). The acid trap was made up of a sponge (diameter, 5 cm; thickness, 1.3 cm) with 5 ml of H_3PO_4–glycerol solution (40 ml glycerol, 50 ml H_3PO_4 acid, and 910 ml deionized water) placed inside the lower part of the column top lid. The acid traps were installed on Day 0 after all treatments were applied to soil. All NH_3 traps were exchanged with fresh ones on Days 1, 2, 3, 5, 7, 9, 11, 13, 17, 21, and 25. Each used trap was thoroughly rinsed in 2 M KCl solution and squeezed several times to extract the solution. The collected extracts were analyzed for NH_4–N using a flow injection method (Ahmed, Stalikas, Tzouwara-Karayanni, Karayannis, & Veltsistas, 1997). Cumulative NH_3 loss was calculated by summing NH_4–N across all collection dates. Cumulative NH_3 loss was converted to kg N ha^{-1} by multiplying the total volatilization loss and the given soil surface area.

Nitrous oxide emissions were measured by collecting gas samples through the septum port on the upper terminal lid. Gas samples were collected on alternate days (Days 1, 3, 5, 7, 9, 11, 13, 15, 17, 19, 21, 23, 25, 27, and 29). During the N_2O sampling period, the NH_3 trap was removed from the column, which gave a headspace of 315 cm^3. Gas samples were collected at 0, 10, and 20 min using a 12-ml syringe. The 0-min samples were collected before closing the lid. At each sampling, gas was transferred to a 10-ml glass sample vial (Wheaton). Samples were analyzed for N_2O concentrations with a gas chromatograph (450-GC, Varian) using an electron capture detector. The N_2O concentration values were converted to mass per volume using the universal gas law equation. Daily gas flux rates (mg m^{-2} min^{-1}) were calculated as the linear or quadratic change in headspace N_2O concentration over time (Wagner, Reicosky, & Alessi, 1997) based on regression analysis with the highest r^2 value. Cumulative N_2O emissions (kg N ha^{-1}) were determined by integrating daily N_2O fluxes using the trapezoidal integration method (Dunmola, Tenuta, Moulin, Yapa, & Lobb, 2010).

An attempt was made to collect column leachate on each day after water addition. On each collection date, suction with a 0.25-horsepower air motor (Model 1603007402, Bluffton Motor Works) was applied to the bottom lid of the column to facilitate drainage of water collected at the bottom of soil column through a porous ceramic plate (Peng et al., 2015). Leachate samples were frozen until analysis for NO_3–N using a flow injection method (Ahmed et al., 1997). The total amount of NO_3–N leached in each treatment was calculated by multiplying NO_3–N concentration with leachate volume and summing over collection dates.

All samplings were done in the morning (8:00 a.m.–12:00 p.m.). At the end of the experiment, the porous ceramic plate was removed from the bottom of the soil column, and soil was divided into 6-cm increments. For each increment, 10 g of soil was collected for determination of GWC, and the remaining soil was analyzed for NH_4–N and NO_3–N concentrations. Soil residual inorganic N was calculated as the sum of NH_4–N and NO_3–N concentrations across all soil increments for each column.

2.2 | Data analysis

The N losses via NH_3 volatilization, NO_3–N leaching, and N_2O emissions and soil residual N in unfertilized treatment (C0N0) were subtracted from those in fertilized treatments and divided by the amount of UAN-N applied (i.e., 39.5 mg N) to estimate those losses per applied UAN-N. Fertilizer N recovery (FNR) was estimated by two methods. Equation 1 represents the "N difference" method, where N losses and residual N at the end of the experiment in control treatment (C0N0) were subtracted from those in fertilized treatment to estimate FNR based on "N difference" method (FNR_{CTRL}) (adapted from Mahal et al. [2019]). Equation 2 estimated FNR based on the initial extractable N (FNR_{ResN}), which accounted for initial extractable N at the beginning of the experiment (adapted from Li, Hu, Delgado, Zhang, & Ouyang, 2007).

$$FNR_{CTRL} = \frac{(\text{N loss}_{\text{Treatment}} - \text{N loss}_{\text{C0N0}}) + (\text{Soil residual N}_{\text{Treatment}} - \text{Soil residual N}_{\text{C0N0}})}{\text{Applied N}} \times 100 \tag{1}$$

$$\mathrm{FNR_{ResN}} = \frac{\mathrm{N\,loss_{Treatment}} + \mathrm{Soil\,residual\,N_{Treatment}}}{(\mathrm{Applied\,N} + \mathrm{Initial\,extractable\,N})} \times 100 \quad (2)$$

The effects of treatment and soil on dependent variables' cumulative values were tested using the PROC MIXED procedure in SAS, with treatment, soil, and their interaction as the fixed effects and rep as random effect (Littell, Milliken, Stroup, Wolfinger, & Schabenberger, 2006; SAS, 2015). When main or interaction effects were significant, means were separated by the LSD test (Littell et al., 2006). Ammonia volatilization and N_2O emissions data were analyzed using repeated measures in ANOVA to determine the differences among treatments by sampling dates. Statistical significance was evaluated at $P < .05$ unless otherwise stated.

3 | RESULTS

3.1 | Ammonia volatilization

Daily NH_3 volatilization loss with the C4N1 treatment was higher than with other treatments in the first 10 acid trap sample collection dates ($n = 12$) in loamy soil (Figure 1a). The same was true for sandy loam on five different sampling dates (Figure 1b). After Day 17, all treatments showed no or minimal volatilization loss in both soil types. In fertilized treatments, all daily NH_3 losses were >2% of applied N and occurred within the first 2 wk of the experiment, and losses were >1% by the third week in both soil types.

Cumulative NH_3 loss across treatments ranged from 0.2 to 9.1 mg (equivalent to 1.0–46.4 kg N ha^{-1}) in loam and from 0.2 to 6.9 mg (equivalent to 1.0–35.2 kg N ha^{-1}) in sandy loam soils. There was a significant treatment × soil interaction effect on cumulative NH_3 loss and cumulative NH_3 loss per applied N (Tables 1 and 2). Compared with C0N1, cumulative NH_3 loss (per applied N) was significantly lower for C1N1, C2N1, and C3N1 in loam soil and for C1N1 and C2N1 in sandy loam soil (Table 2). The C3N1 and C4N1 in sandy loam and C4N1 in loam increased NH_3 loss (per applied N) compared with C0N1. The C0N0 and C4N1 had minimal NH_3 losses in both soil types (Figure 1). Among fertilized treatments (C0N1, C1N1, C2N1, C3N1, and C4N1), cumulative NH_3 loss per applied N ranged from 3.2 to 22.3% in loam and from 6.6 to 16.8% in sandy loam soils.

3.2 | Nitrous oxide emissions

Daily N_2O fluxes varied from 0 to 0.4 mg m^{-2} h^{-1} in loam and were 0.3 mg m^{-2} h^{-1} in the sandy loam soil across treatments throughout the experiment (Figure 2). Variability in daily N_2O fluxes was high among replications in both loam (coefficient of variance [CV], 32.1–166.1%) and sandy loam (CV, 12.1–176.2%). Of the 15 sampling dates, C0N1 had the highest daily N_2O flux on the final sampling date in loam and on Days 7 and 9 in sandy loam. Control treatments always had minimal N_2O fluxes in both soil types.

Cumulative N_2O emissions differed by treatment but did not differ by soil type or their interaction (Table 1). Emissions were greater in fertilized treatments compared with unfertilized treatments at $P < .001$. Cumulative N_2O emissions among fertilized treatments were not different. Averaged cumulative N_2O emissions in fertilized treatments were 0.7 kg N ha^{-1} in both soil types and 0.03 and 0.05 kg N ha^{-1} in unfertilized loam and sandy loam, respectively (Supplemental Figure S6). Among fertilized treatments (C0N1, C1N1, C2N1, C3N1, and C4N1), cumulative N_2O emissions per applied N ranged from 0.1 to 0.5% in loam and from 0.1 to 0.4% in sandy loam soils.

3.3 | Nitrate leaching

In loam, one leaching event occurred on Day 29 after N fertilization across all treatments. In contrast, three leaching events occurred in sandy loam (Days 20, 21, and 29), with 44.3% of the total NO_3–N leaching observed on Day 29 (Figure 3).

There was a significant treatment × soil interaction effect on cumulative NO_3–N leaching (Tables 1 and 2). Cumulative NO_3–N leaching was consistently greater for all fertilized treatments in sandy loam than in loam (Table 2). Averaged across all treatments, cumulative NO_3–N leaching was almost fourfold greater for sandy loam (17.6×10^{-3} g) than for loam (4.3×10^{-3} g) (Table 1).

Among fertilized treatments, cumulative NO_3–N leaching per applied N was higher in sandy loam (32.4%) than in loam (2.6%) (Table 1). In sandy loam, C3N1 had lower NO_3–N leaching (16.9×10^{-3} g or 21.1% of applied N) than C0N1 (24.3×10^{-3} g or 39.9% of applied N) (Table 2).

3.4 | Soil residual mineral nitrogen and fertilizer nitrogen recovery

There was a significant treatment × soil interaction effect on soil residual mineral N throughout the column (Table 1). Control treatments (C0N0 and C4N0) had lower soil residual mineral N than fertilized treatments in both soil types (Table 2). Among fertilized treatments, soil residual mineral N was similar in sandy loam but was significantly lower in C4N1 (26.4×10^{-3} g or 49.8% of applied N) than in the other treatments in loam (Table 2).

When separated by depth, soil residual N was greater in fertilized treatments than in the control treatments at 18–24 cm

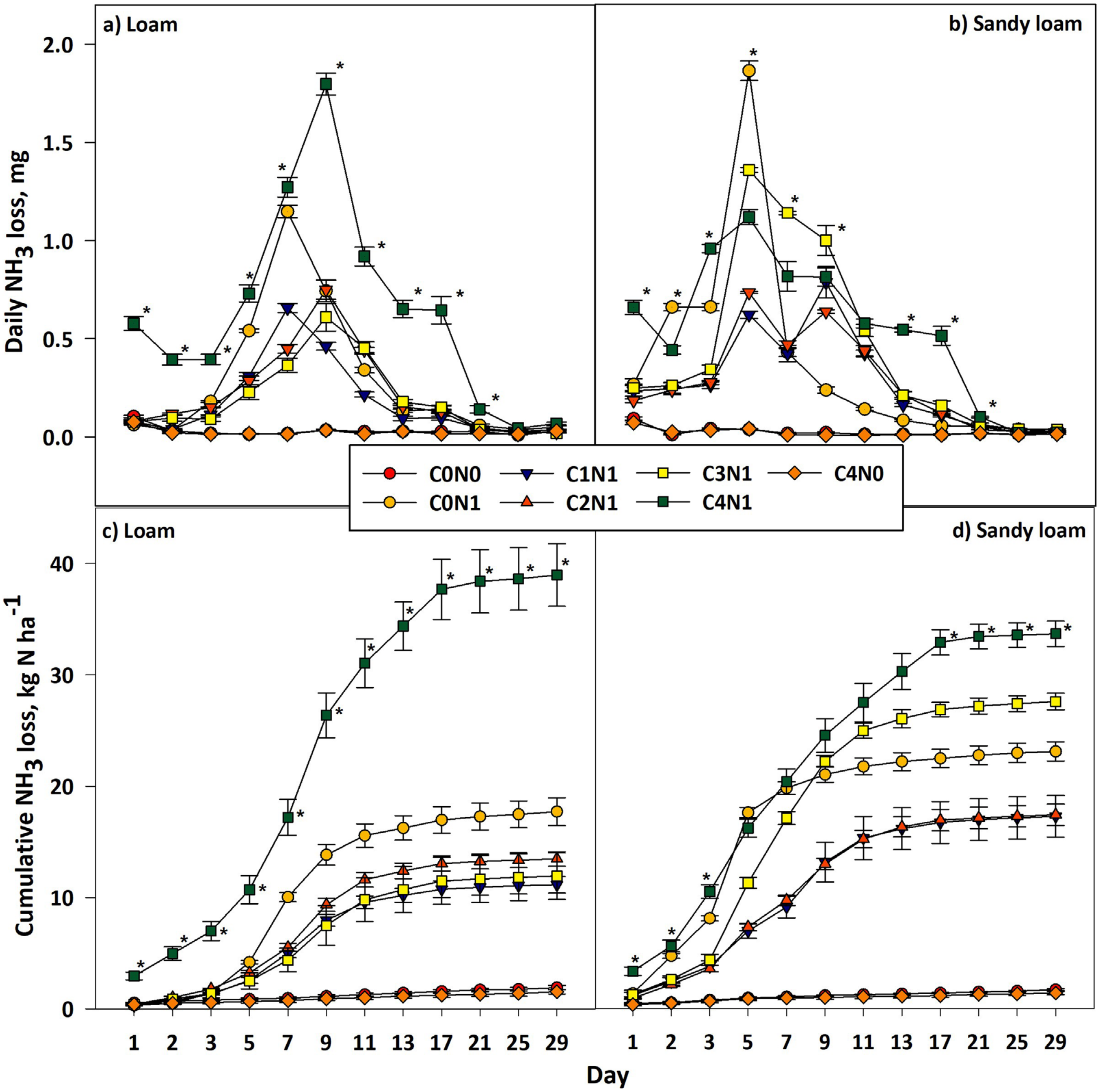

FIGURE 1 Daily and cumulative NH_3 volatilization loss (mean ± SE) with different treatments in (a, c) loam and (b, d) sandy loam soils. C0N0, no char and no urea ammonium nitrate (UAN); C0N1–C4N1, UAN at 200 kg N ha^{-1} and char at 0, 6.7, 10.1, 13.4, and 26.8 Mg C ha^{-1}, respectively; C4N0, 26.8 Mg C ha^{-1} and no UAN. *Treatment with significantly higher loss than all other treatments on a given sampling day

in both soil types. Fertilized treatments (C0N1, C1N1, C2N1, C3N1, and C4N1) in loam had greater residual N than the control treatments (C0N0 and C4N0) at other depths as well. Soil residual mineral N at 18–24 cm was higher with C1N1 and C3N1 in loam soil than other treatments in both soil types (Figure 4). In loam, C4N1 had lower soil residual N at 18–24 cm than other fertilized treatments. In sandy loam, soil residual N were greater with C3N1 than other treatments except C1N1. Among fertilized treatments in sandy loam, C4N1 and C0N1 had lower soil residual N than others.

There were no significant differences by treatment or soil in fertilizer N recovery (Table 1). The FNR_{CTRL} ranged from 67.6 to 77.3% by soil type and from 69.0 to 74.2% by treatment. The UAN-N applied among fertilized treatments (C0N1, C1N1, C2N1, C3N1, and C4N1) that remained unaccounted ranged from 26.3 to 34.4%. However, FNR_{ResN}

TABLE 1 Analysis of variance results with means for different dependent variables as affected by char, soil, and their interaction

	NH_3 volatilized		N_2O emissions		NO_3–N leached		Soil residual mineral N		FNR_{CTRL} [b]	FNR_{ResN} [c]
Treatment[a]	g (10^{-3})	% per applied N	g (10^{-3})	% per applied N	g (10^{-3})	% per applied N	g (10^{-3})	% per applied N	%	
C0N0	0.4	–	0.01b	–	6.1	–	6.2	–	–	–
C0N1	4.0	9.2	0.12a	0.28	14.3	20.9	22.8	42.1	72.5	97.3
C1N1	2.8	6.2	0.11a	0.28	12.2	15.4	26.4	51.2	73.1	97.8
C2N1	3.0	6.8	0.15a	0.34	14.1	20.4	24.6	46.7	74.2	95.0
C3N1	3.9	8.9	0.10a	0.25	10.8	11.9	25.1	48.0	69.0	94.0
C4N1	7.1	17.1	0.15a	0.36	13.6	19.1	20.9	37.1	73.7	98.3
C4N0	0.3	–	0.01b	–	5.4	–	7.0	–	–	–
Significance	***	***	***	NS	***	NS	***	NS	NS	NS
Soil										
Loam	2.7	8.7	0.10	0.32	4.3	2.6b	25.4	66.1a	77.3	96.2
Sandy loam	3.4	11.0	0.09	0.28	17.6	32.4a	12.5	23.9b	67.6	96.7
Significance	***	***	NS	NS	***	***	***	***	NS	NS
Treatment × soil	***	***	NS	NS	***	NS	***	NS	NS	NS

Note. Means in a column followed by same lowercase letter are not significantly different. When interaction effect was significant, main effect was not reported.

[a]C0N0, no char and no urea ammonium nitrate (UAN); C0N1–C4N1, UAN at 200 kg N ha^{-1} and char at 0, 6.7, 10.1, 13.4, and 26.8 Mg C ha^{-1}, respectively; C4N0, 26.8 Mg C ha^{-1} and no UAN.

[b]Fertilizer N recovery based on "N difference" method.

[c]Fertilizer N recovery based on the initial extractable N.

*Significant at the .05 probability level.

**Significant at the .01 probability level.

***Significant at the .001 probability level. NS, not significant.

TABLE 2 Interaction effect of treatment and soil on cumulative NH_3 volatilized, NO_3–N leached, and soil residual N

	NH_3 volatilized		NO_3–N leached		Soil residual N	
	Loam	Sandy loam	Loam	Sandy loam	Loam	Sandy loam
Treatment[a]	g (10^{-3})					
C0N0	0.4g	0.3g	3.6cd	8.5c	6.7e	5.6e
C0N1	3.5e	4.5d	4.4cd	24.3a	33.3a	12.4cd
C1N1	2.2f	3.4e	1.5d	22.9ab	36.6a	16.2c
C2N1	2.6f	3.4e	5.8cd	22.5ab	35.1a	14.2c
C3N1	2.3f	5.4c	4.6cd	16.9b	32.8a	17.5c
C4N1	7.6a	6.6b	7.0cd	20.2ab	26.4b	15.2c
C4N0	0.3g	0.3g	3.2cd	7.7cd	7.0de	6.4e

Note. Means for each variable followed by same lowercase letters are not significantly different.

[a]C0N0, no char and no urea ammonium nitrate (UAN); C0N1–C4N1, UAN at 200 kg N ha^{-1} and char at 0, 6.7, 10.1, 13.4, 26.8 Mg C ha^{-1}, respectively; C4N0, 26.8 Mg C ha^{-1} and no UAN.

ranged from 94.0 to 98.3% in treatments and from 96.2 to 96.7% by soil type (Table 1).

4 | DISCUSSION

4.1 | Ammonia volatilization

Ammonia volatilization loss observed in this study aligned with other studies that reported NH_3 losses from 8 to 13% (Ma et al., 2010a; Peng et al., 2015; Vaio et al., 2008). Char addition did not enhance or suppress NH_3 volatilization in unfertilized treatments. Fertilization is the major source for NH_3 volatilization loss, as evidenced by a positive correlation between NH_3 volatilization and N fertilization reported in Jantalia et al. (2012) and Jones, Brown, Engel, Horneck, and Olson-Rutz (2013).

The higher clay content and CEC in loam than in sandy loam promoted better retention of NH_4 and subsequently reduced NH_3 loss in loam compared with sandy loam in this

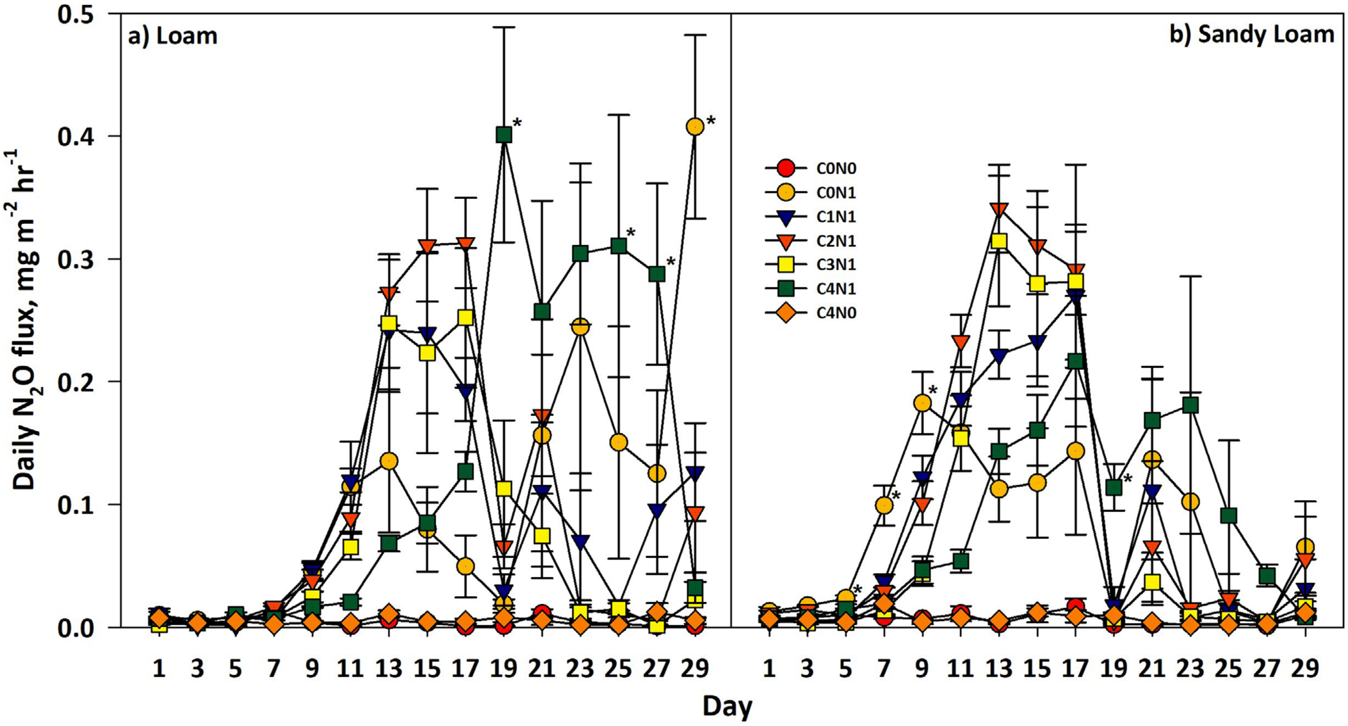

FIGURE 2 Daily N_2O flux (mean ± SE) with different treatments in (a) loam and (b) sandy loam soils. C0N0, no char and no urea ammonium nitrate (UAN); C0N1–C4N1, UAN at 200 kg N ha^{-1} and char at 0, 6.7, 10.1, 13.4, and 26.8 Mg C ha^{-1}, respectively; C4N0, 26.8 Mg C ha^{-1} and no UAN. *Treatment with significantly higher loss than all other treatments on a given sampling day

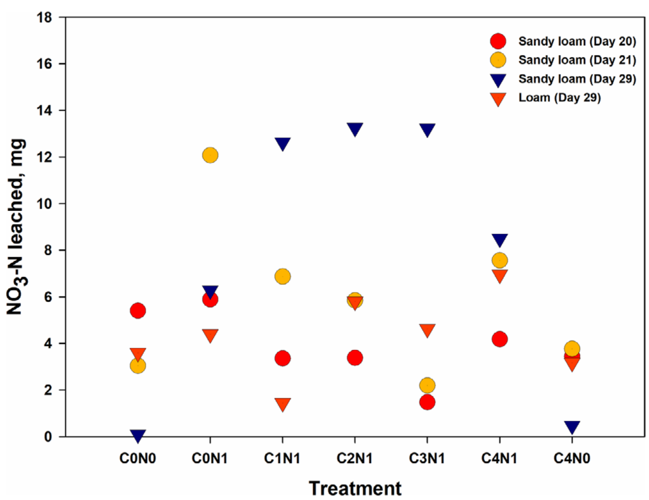

FIGURE 3 Amount of NO_3–N leached (mean; $n = 4$) with different treatments in loam and sandy loam soils at different leaching events. C0N0, no char and no urea ammonium nitrate (UAN); C0N1–C4N1, UAN at 200 kg N ha^{-1} and char at 0, 6.7, 10.1, 13.4, and 26.8 Mg C ha^{-1}, respectively; C4N0, 26.8 Mg C ha^{-1} and no UAN. Leaching events occurred on Days 20, 21, and 29 after fertilization for sandy loam and on Day 29 for loam

study. In addition, a higher sand content would enhance the loss of NH_3 in sandy loam (McDowell et al., 1958).

Reduction in NH_3 volatilization observed at lower char rates in both soil types was likely from increased physisorption due to the high surface area (82.1 m^2 g^{-1}) and the high CEC (46.9 meq 100 g^{-1}) of char. The surface area of char exceeds that of clay-sized particles (Qi & Zhang, 2015) by one or two orders of magnitude and exceeds that of sand particles by three or four orders of magnitude. These results suggest that char functions more like biochar from various sources that have been reported to capture NH_3 and reduce NH_3 volatilization loss (Steiner et al., 2010; Taghizadeh-Toosi, Clough, Sherlock, & Condron, 2012). However, the beneficial effect of high-C products, such as char and biochar, in reducing NH_3 loss depends on their sources, production conditions, contaminments and quality, and application rates (Ding et al., 2016; Steiner et al., 2008).

Soil pH is another important factor for retention or release of NH_4/NH_3 in the soil. At pH below 7.5, NH_4 is the predominant form, rather than volatile NH_3 (Fan et al., 1993). As pH increases above 7.5, the NH_3 form quickly becomes dominant and is susceptible to loss via volatilization (Behera, Sharma, Aneja, & Balasubramanian, 2013). The initial pH of sandy loam in this study was 7.7, which is above the 7.5 pH threshold for NH_3 volatilization, whereas the loam soil had a pH of 7.2, which is slightly below this threshold. The pH of the char was 7.6, and char contained 19% calcium carbonate. Calcium carbonate aids in increasing soil alkalinity, and hydrolysis of urea to form NH_4 also raises the pH (Jones et al., 2013). Depending on the nature and composition of CCRs, they could be useful to increase or buffer soil pH (Elseewi, Bingham, & Page, 1978a; Elseewi, Bingham, & Page, 1978b;

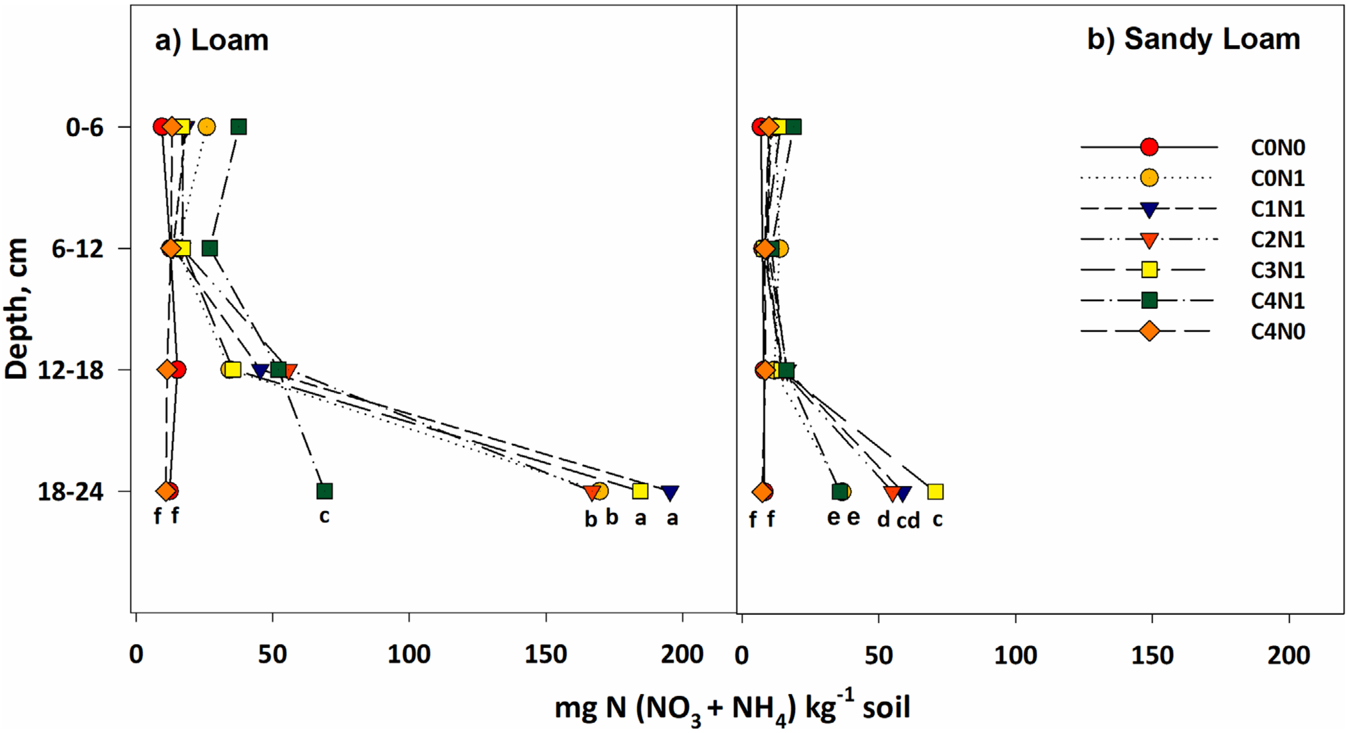

FIGURE 4 Soil residual N (mean; $n = 4$) the 0- to 24-cm depth at different treatments in (a) loam and (b) sandy loam soils. C0N0, no char and no urea ammonium nitrate (UAN); C0N1–C4N1, UAN at 200 kg N ha^{-1} and char at 0, 6.7, 10.1, 13.4, and 26.8 Mg C ha^{-1}, respectively; C4N0, 26.8 Mg C ha^{-1} and no UAN. Means at 18–24 cm with different letters across both soil types are significantly different at $P < .05$

Phung, Lund, & Page, 1978). There could have been a considerable soil alkalization effect with higher char rates that counteracted and exceeded physiosorption benefits of char.

4.2 | Nitrous oxide emissions

The average N_2O emissions rate of 0.7 kg N ha^{-1} from fertilized treatments in this study is comparable to the 0.6 kg N ha^{-1} emission rate from UAN at 150 kg N ha^{-1} in a 28-d field study in eastern Canada (Ma et al., 2010b). In this study, a considerable amount of N moved down the soil profile and/or leached, and char addition would have only facilitated that downward N movement (Basu et al., 2009). A slight increase in N_2O emissions in loam soil compared with sandy loam (Table 1) could be related to anaerobic conditions at some pockets in loam soil, which promotes denitrification (Weier, Doran, Power, & Walters, 1993).

A previous laboratory incubation study documented that N_2O emissions may vary by soil texture (Harrison-Kirk, Beare, Meenken, & Condron, 2013), but no significant differences in N_2O emissions by soil types were found in our study. Nitrous oxide emissions are primarily driven by N fertilization (Maharjan, Venterea, & Rosen, 2014; Shcherbak, Millar, & Robertson, 2014), as evidenced by greater emissions in fertilized than unfertilized treatments in this study. The high variability in daily N_2O fluxes among laboratory replicates, which is likely be larger under field conditions, was one reason for the nonsignificant differences and should be kept in mind for evaluation of N losses from agricultural systems because it points toward a highly dynamic pathway. Johnson and Welch (1939) suggested 33% as permissible upper fiducial limit of CV. Although the acceptable range of CV may vary among experiments, the high CV observed in daily fluxes in this study failed to detect differences in treatment means (Patel, Patel, & Shiyani, 2001). Another potential pitfall in this study could be the small headspace used for gas sampling, which reduces minimum detectable flux (De Klein and Harvey, 2012).

4.3 | Nitrate leaching

The contrasting effect of C3N1 and C0N1 in sandy loam with respect to NH_3 loss and NO_3–N leaching underscores the need to account for all possible pathways of N losses in our mitigation efforts. The lower NO_3–N leaching loss in C3N1 than in C0N1 is due to greater soil mineral residual N at the lower bottom of the column (18–24 cm depth) and greater NH_3 loss in C3N1 than in C0N1. When there are multiple possible pathways for loss, as is the case with mineral N, an effort to reduce N loss via a particular pathway may be undermined or even outweighed by loss via other pathway(s) (Lam, Suter, Mosier, & Chen, 2016).

The effect of high-C-content amendments on NO_3–N leaching depends on complex physical, chemical, and biological processes. It has been suggested that leaching of soil NO_3–N depends on the ability of biochar to retain NO_3–N and NH_4–N or on the inhibition of nitrification by clay particles (Clough, Condron, Kammann, & Müller, 2013; Liu et al., 2017). Some biochar studies have found decreased NO_3–N leaching depending on fertilizer type, soil type, and leaching conditions, but other studies showed inconsistent effects of biochar on leaching (Fidel, Laird, & Spokas, 2018; Haider, Steffens, Moser, Müller, & Kammann, 2017; Sika & Hardie, 2014).

Ventura, Sorrenti, Panzacchi, George, and Tonon (2013) observed a reduction in NO_3–N leaching only in the second year after biochar application, suggesting an increase in biochar sorption properties over time, possibly due to the oxidation and interaction of biochar and soil particles and an increase in the adsorbing surface due to particle fragmentation with aging (Hagemann et al., 2017; Singh, Hatton, Singh, Cowie, & Kathuria, 2010). In contrast, Gronwald, Don, Tiemeyer, and Helfrich (2015) observed that the adsorption capacity of biochar decreased by 60–80% to less or observed no NO_3/NH_4–N adsorption after 7 mo of aging in the field compared with the fresh char. A similar trend of decreasing adsorption capacity with biochar from beetroot chips was reported from a laboratory study on loam soil (Bargmann, Martens, Rillig, Kruse, & Kücke, 2014). Possible reasons for decreased adsorption capacity over time can be binding sites of biochar being blocked with organic matter or mineral particles and microbial degradation with subsequent possible changes in surface properties (Cheng, Lehmann, & Engelhard, 2008). In this study, a leaching event was observed on Day 29 after fertilization in loamy soil. The later and lower NO_3–N leaching observed in fertilized loam than in sandy loam in this study may be due to a lower water infiltration rate and greater nutrient retention in loamy soil because of greater clay and OM content (Lehmann & Schroth, 2003). Long-term evaluation is required to understand how char properties might change and affect soil NO_3–N leaching over time.

4.4 | Soil residual mineral nitrogen and fertilizer nitrogen recovery

Lower soil residual mineral N at a depth of 18–24 cm and subsequently lower residual mineral N in the whole soil column with C4N1 compared with other fertilized treatments in loam soil could be the result of higher NH_3 volatilization loss (cumulative loss of 7.6×10^{-3} g N or 17.1% of applied N) (Figure 1) or a slightly higher NO_3–N leaching loss (Table 1).

In all fertilized treatments, most of N moved down the profile and accumulated at the lower soil layers of the columns 30 d after N addition. This suggests the movement of NO_3–N down the soil profile with water (Bahmani, Nasab, Behzad, & Naseri, 2009; Pierzynski, Vance, & Sims, 2005). Previous research documented that 25.4 mm of irrigation or rainfall can transport soil NO_3–N to 150–200 mm in a loamy sand (Endelman, Keeney, Gilmour, & Saffigna, 1974). During the 30-d experiment, 100.8 mm of water was added. In the case of sandy loam soil, N moved down the profile and leached out of the column; therefore, residual mineral N was overall lower in sandy loam than in loam across fertilized treatments, including C4N1.

The FNR_{CTRL} was much smaller than FNR_{ResN} (Table 1). The FNR_{CTRL} estimate assumes that fertilizer N enhances OM mineralization (Khan, Mulvaney, Ellsworth, & Boast, 2007; Robertson et al., 2013). However, inorganic N inputs can also decrease OM mineralization by decreasing the decomposition of energy-poor OM substrates that are mineralized solely to access N-containing compounds (Craine, Morrow, & Fierer, 2007; Moorhead & Sinsabaugh, 2006). Particularly, in the current study, no crops were grown, and therefore there was no OM to mineralize to make up for potential N deficiency. In a laboratory incubation study with no crops involved, Mahal et al. (2019) demonstrated that fertilizer N suppressed OM mineralization. In contrast, Kaleeem Abbasi, Mahmood Tahir, Sabir, and Khurshid (2015) reported that control soil without amendment released a maximum of 30.9 mg N kg^{-1} soil on Day 28 compared with 13.7 mg kg^{-1} at Day 0 at 25°C and 58% water filled pore space under laboratory conditions, showing a substantial release of N into the mineral N pool. The wide variation reported in the N mineralization from soils with or without fertilizer N can be affected by applied N rate (Cahill, Osmond, Crozier, Israel, & Weisz, 2007), soil temperature and moisture (Deenik, 2006), amount and type of clay in soil (Breland, 1994; Deenik, 2006). In the current study, mineralization under different treatments were not measured and the long-term effect of char-C in soil N mineralization/immobilization is yet to be explored. Irrespective of the methods of estimating FNR, it did not vary by treatments or soil. However, the differences in FNR_{CTRL} and FNR_{ResN} observed in this study underscores the implications of different methods used in calculating fertilizer N recovery or use efficiency (Mahal et al., 2019) and a critical role that soil OM mineralization might play in soil N availability and N use efficiency.

5 | CONCLUSION

In many countries, CCRs have not been properly utilized and still considered as waste products. Benefits of decreasing NH_3 volatilization loss were observed with optimum rates of char addition in both coarse and fine-textured soils. There were no adverse effects of adding char on leaching losses or N_2O emissions. Field research is warranted to evaluate the potential use of char and other similar high C content by-products to improve N management. Further evaluation is warranted to investigate the possible adverse effects of pesticide/herbicide sorption and potential trace metal accumulation in soil, crop tissue, or grains before recommending char for agricultural use.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

ORCID

Dinesh Panday https://orcid.org/0000-0001-8452-3797
Arindam Malakar https://orcid.org/0000-0001-6704-8891
Bijesh Maharjan https://orcid.org/0000-0002-4728-7956

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

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