



Leaching losses of dissolved organic carbon and nitrogen from agricultural soils in the upper US Midwest

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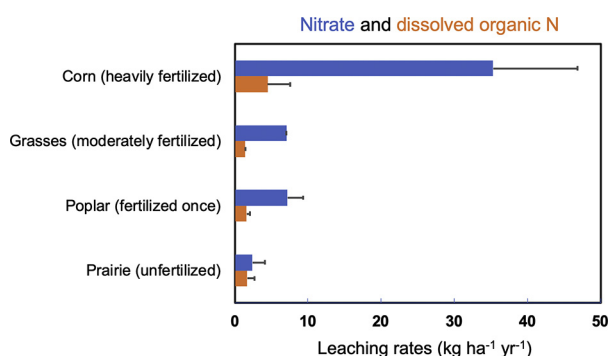
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HIGHLIGHTS

- Organic carbon leaching was unaffected by nitrogen fertilization, but it was greater in poplar than in the herbaceous crops.
- Nitrogen was leached mainly as nitrate, with the balance lost mostly as organic N and relatively little as ammonium.
- Leaching of organic nitrogen increased with N fertilization.
- Perennial cropping systems that were moderately fertilized leached less nitrogen than more heavily fertilized corn.

GRAPHICAL ABSTRACT



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ABSTRACT

Leaching losses of dissolved organic carbon (DOC) and nitrogen (DON) from agricultural systems are important to water quality and carbon and nutrient balances but are rarely reported; the few available studies suggest linkages to litter production (DOC) and nitrogen fertilization (DON). In this study we examine the leaching of DOC, DON, NO_3^- , and NH_4^+ from no-till corn (maize) and perennial bioenergy crops (switchgrass, miscanthus, native grasses, restored prairie, and poplar) grown between 2009 and 2016 in a replicated field experiment in the upper Midwest U.S. Leaching was estimated from concentrations in soil water and modeled drainage (percolation) rates. DOC leaching rates ($\text{kg ha}^{-1} \text{ yr}^{-1}$) and volume-weighted mean concentrations (mg L^{-1}) among cropping systems averaged 15.4 and 4.6, respectively; N fertilization had no effect and poplar lost the most DOC (21.8 and 6.9, respectively). DON leaching rates ($\text{kg ha}^{-1} \text{ yr}^{-1}$) and volume-weighted mean concentrations (mg L^{-1}) under corn (the most heavily N-fertilized crop) averaged 4.5 and 1.0, respectively, which was higher than perennial grasses (mean: 1.5 and 0.5, respectively) and poplar (1.6 and 0.5, respectively). NO_3^- comprised the majority of total N leaching in all systems (59–92%). Average NO_3^- leaching ($\text{kg N ha}^{-1} \text{ yr}^{-1}$) under corn (35.3) was higher than perennial grasses (5.9) and poplar (7.2). NH_4^+ concentrations in soil water from all cropping systems were relatively low ($<0.07 \text{ mg N L}^{-1}$). Perennial crops leached more NO_3^- in the first few years after planting, and markedly less after. Among the fertilized crops, the leached N represented 14–38% of the added N over the study period; poplar lost the greatest proportion (38%) and corn was intermediate (23%).

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Requiring only one third or less of the N fertilization compared to corn, perennial bioenergy crops can substantially reduce N leaching and consequent movement into aquifers and surface waters.

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1. Introduction

Dissolved organic matter leached from litter and soils into water percolating downward can play an important role in biogeochemical processes within the soil profile and in downstream ground and surface waters (Qualls and Haines, 1991). Dissolved organic matter is an energy source for heterotrophic microbes and may control the rate of denitrification in the unsaturated zone and in downstream flow paths, which in turn affects nitrate (NO_3^-) concentrations in groundwater and surface waters (Brye et al., 2001; Rivett et al., 2008). In addition, dissolved organic matter can form complexes with trace metals, thereby increasing their bioavailability (Hernández-Soriano and Jiménez-López, 2015).

Studies in non-agricultural ecosystems have shown that leaching losses represent a significant export of carbon (C) and nitrogen (N) (Campbell et al., 2000; Jones et al., 2004; Dijkstra et al., 2007; Kindler et al., 2011; Camino-Serrano et al., 2016; Thieme et al., 2019). Whether this is true in agricultural systems (agroecosystems) is not well established because studies often focus on inorganic N and do not measure dissolved organic carbon (DOC) and dissolved organic nitrogen (DON) (Filep and Rékási, 2011), although there is evidence from a few locations that DOC leaching can be a significant carbon loss pathway in agricultural systems (Kindler et al., 2011; Manninen et al., 2018). Agroecosystems are at least as productive as the native ecosystems they replace, but much of the biomass is often removed in harvest, thereby reducing the availability of detrital organic matter for leaching after the plants senesce. Compared to annual crops like corn, productive perennial crops may have more turnover of leaf and root biomass over the growing season prior to harvest (e.g., Schläpfer and Ryser, 1996), and thus may leach more DOC and DON.

Nitrate has been identified as the main form of N leached from many fertilized agroecosystems (Basso and Ritchie, 2005; Robertson and Saad, 2011; Hussain et al., 2019). Because of its high mobility in soils, NO_3^- moves in percolating water more readily than NH_4^+ and often reaches surface water and groundwater in high concentrations, potentially degrading drinking water quality as well as contributing to eutrophication of downstream waters. Both NO_3^- and DON leaching from agricultural soils can contribute to eutrophication in water bodies receiving groundwater or tile drain discharge (van Kessel et al., 2009; Baron et al., 2013). Moreover, studies suggest that DON leaching losses could be important in ecosystem N budgets. Although compared to NO_3^- there have been relatively few studies of DON in agroecosystems (Murphy et al., 2000), the relative importance of DON to total N leaching losses has occasionally been reported and shows large variability. A review by van Kessel et al. (2009), which found only 16 studies that reported leaching losses of both DON and total dissolved nitrogen (TDN), concluded that DON averaged 28% of the TDN losses from a diverse set of agroecosystems.

Comparative studies of DON leaching in relation to fertilization in various annual and perennial cropping systems in the same location are lacking, but based on a number of experimental N additions to forests and the fewer studies that have examined fertilized agroecosystems, leaching losses of DON likely increase with N fertilization (Gundersen et al., 1998; Siemens and Kaupenjohann, 2002; van Kessel et al., 2009). Corn (*Zea mays* L.; a.k.a. maize), an annual crop grown on ~70 million ha throughout the US Midwest (USDA-ERS, 2015), is heavily fertilized and is a major source of N pollution of ground and surface waters (McIsaac et al., 2010; Smith et al., 2013; Padilla et al., 2018; Wang et al., 2019). Corn systems typically lose 15–65% of the applied N to leaching as NO_3^- (Randall and Mulla, 2001; Basso et al., 2016;

Hussain et al., 2019). The available studies comparing NO_3^- to DON leaching in corn systems suggest that about 1–25% of applied N is lost as DON (Shuster et al., 2003; Jiao et al., 2004; Salazar et al., 2019).

The main goals of this study are to quantify the leaching losses of DOC and DON and compare them to losses of N as NO_3^- and NH_4^+ among diverse annual and perennial cropping systems grown in a replicated experimental design. This work builds on our previous study of NO_3^- leaching from these systems (Hussain et al., 2019) by adding estimates of leaching of DOC, DON, and NH_4^+ , all of which are less often studied in agroecosystems than NO_3^- leaching. We measured concentrations in soil water sampled from below the root zone in five perennial systems (miscanthus, switchgrass, native grasses, restored prairie, and hybrid poplar) and in corn. Leaching losses were estimated by combining measured concentrations with modeled drainage rates. All systems were no-till crops grown in close proximity on a well-drained sandy loam soil in the humid temperate climate of southwestern Michigan. We estimated leaching losses over seven crop-years from 2009 to 2016, a period spanning the establishment and stabilization of the perennial cropping systems. We expected larger NO_3^- leaching losses from the fertilized annual corn crop and that these might correspond with higher DON losses, whereas DOC losses might be higher from the perennial cropping systems.

2. Materials and methods

2.1. Experimental site and agronomic details

The Bioenergy Cropping System Experiment (BCSE) is part of the Great Lakes Bioenergy Research Center (www.glbrc.org) and Long Term Ecological Research (LTER) program (www.lter.kbs.msu.edu). The BCSE is located at the W.K. Kellogg Biological Station (KBS) (42.3956° N, 85.3749° W and 288 m above sea level), 25 km from Kalamazoo in southwestern Michigan, USA. Soils are Typic Hapludalfs developed on glacial outwash (Thoen, 1990; Crum and Collins, 1995) with intermixed loess (Luehmann et al., 2016), and are sandy loams near the surface (sand content usually >40%) with higher % sand at depth (usually >80% below 50 cm) (Table S1). The site is well-drained with the water table approximately 12–14 m below the surface. The climate is humid temperate with a mean annual air temperature of 10.1 °C and annual precipitation of 1005 mm, 511 mm of which falls between May and September (means for 1981–2010: NCDC, 2013).

The BCSE was established in 2008 on land previously used for row crop agriculture for many decades. The experimental design consists of five randomized blocks, and each block consists of ten cropping system plots of 28 × 40 m (Fig. S1). The ten cropping systems include (1) continuously grown no-till corn; (2) switchgrass, *Panicum virgatum* L. (var. Cave-in-Rock); (3) giant miscanthus, *Miscanthus × giganteus* Greef & Deuter ex Hodkinson & Renvoize; (4) a five species native grass assemblage (*Andropogon gerardii* Vitman, *Elymus canadensis* L., *Panicum virgatum* (var. Southlow), *Schizachyrium scoparium* [Michx.] Nash, and *Sorghastrum nutans* [L.] Nash, four of which are C₄ grasses; (5) an 18-species restored prairie with grasses and forbs; and (6) hybrid poplar trees (*Populus nigra* × *P. maximowiczii* 'NM6') with a low-stature herbaceous understory. Species composition in the restored prairie is listed in Table S2. All perennial grasses and poplar were planted between May and June 2008, although switchgrass, native grasses, and restored prairie were re-planted in July 2009 due to poor establishment after a heavy rainfall. All crops were grown without irrigation and under no-till management.

Application of N fertilizer to different crops followed typical local practices (Table 1). In this paper, leaching data are presented based on crop-years (i.e., beginning with spring planting for annual crops or leaf emergence for perennials), and as a result, corresponding N fertilizer rates vary from year to year depending on the planting or emergence dates. Depending on the year, corn was fertilized at 150–170 kg N ha⁻¹ yr⁻¹ as ammonium sulfate in three splits, viz., the first split either prior to planting or at the time of planting depending on the year, the second split a week later, and the third split in the middle of June. All perennial grasses (except restored prairie) were given 56 kg N ha⁻¹ yr⁻¹ between 2010 and 2015, with an additional 77 kg N ha⁻¹ given to miscanthus in 2009. Hybrid poplar was fertilized once in 2010, the point at which the canopy had closed, with 157 kg N ha⁻¹ as ammonium nitrate following silvicultural practices common to the region (Hansen, 1994). The restored prairie was not fertilized.

Corn was planted in the first week of May. Leaf emergence in the perennials, as indicated by phenology cameras, occurred between mid-April and mid-May. Harvest dates depended on weather, varying between October and November for corn and November and December for perennials. Hybrid poplar was harvested once in the winter of 2013–14. The end of the growing season was observed with phenology cameras unless it was truncated by harvest. Depending on the year, the growing season for corn varied between 160 and 190 days, and the growing seasons for perennials including poplar were a few weeks longer than corn (Fig. S2).

2.2. Soil water sampling and laboratory analysis

Prenart soil water samplers made of Teflon and silica (<http://www.prenart.dk/soil-water-samplers/>) were installed in blocks 1 and 2 of the BCSE (Fig. S1), and Eijkelkamp soil water samplers made of ceramic (<http://www.eijkelkamp.com>) were installed in blocks 3 and 4 (there were no soil water samplers in block 5). All samplers were installed at 1.2 m depth at a 45° angle from the soil surface, approximately 20 cm into the unconsolidated sand of the 2Bt2 and 2E/Bt horizons. Beginning in 2009, soil water was sampled at weekly to biweekly intervals during non-frozen periods (April to November) by applying 50 kPa of vacuum for 24 h, during which water was collected in glass bottles. During the 2009 and 2010 sampling periods we obtained fewer soil water samples from blocks 1 and 2 where Prenart lysimeters were installed. We observed no consistent differences between the two sampler types in concentrations of the analytes reported here.

Depending on the volume of leachate collected, water samples were filtered using either 0.45 µm pore size, 33-mm-dia. cellulose acetate membrane filters when volumes were <50 ml, or 0.45 µm, 47-mm-dia. Supor 450 membrane filters for larger volumes. Samples were analyzed for NO₃⁻, NH₄⁺, total dissolved nitrogen (TDN), and DOC. The NO₃⁻ concentration was determined using a Dionex ICS1000 ion chromatograph system with membrane suppression and conductivity detection; the detection limit of the system was 0.006 mg NO₃⁻ N L⁻¹. The NH₄⁺ concentration in the samples was determined using a Thermo Scientific (formerly Dionex) ICS1100 ion chromatograph system with membrane

suppression and conductivity detection; the detection limit of the system was similar. The DOC and TDN concentrations were determined using a Shimadzu TOC-Vcph carbon analyzer with a total nitrogen module (TNM-1); the detection limit of the system was ~0.08 mg C L⁻¹ and ~0.04 mg N L⁻¹. DON concentrations were estimated as the difference between TDN and dissolved inorganic N (NO₃⁻ + NH₄⁺) concentrations. The NH₄⁺ concentrations were only measured in the 2013–2015 crop-years, but they were always small relative to NO₃⁻ and thus their inclusion or lack of it was inconsequential to the DON estimation.

2.3. Nutrient leaching estimates

Leaching rates were estimated on a crop-year basis, defined as the period from planting or emergence of the crop in the year indicated through the ensuing year until the next year's planting or emergence. For each sampling point, the concentration was linearly interpolated between sampling dates during non-freezing periods (April through November). The concentrations in the unsampled winter period (December through March) were also linearly interpolated based on the preceding November and subsequent April samples.

Solute leaching (kg ha⁻¹) was calculated by multiplying the daily solute concentration in pore-water (mg L⁻¹) by the modeled daily drainage rates (m³ ha⁻¹) from the overlying soil. The drainage rates were obtained using the SALUS (Systems Approach for Land Use Sustainability) model (Basso and Ritchie, 2015). SALUS simulates yield and environmental outcomes in response to weather, soil, management (planting dates, plant population, irrigation, nitrogen fertilizer application, tillage), and crop genetics. The SALUS water balance sub-model simulates surface run-off, saturated and unsaturated water flow, drainage, root water uptake, and evapotranspiration during growing and non-growing seasons (Basso and Ritchie, 2015). Drainage amounts and rates simulated by SALUS have been validated with measurements using large monolith lysimeters at a nearby site at KBS (Basso and Ritchie, 2005). On days when SALUS predicted no drainage, the leaching was assumed to be zero. The volume-weighted mean concentration for an entire crop-year was calculated as the sum of daily leaching (kg ha⁻¹) divided by the sum of daily drainage rates (m³ ha⁻¹). Weather data for the model were collected at the nearby KBS LTER meteorological station (lter.kbs.msu.edu).

2.4. Data analyses

One-way analysis of variance (ANOVA) was conducted to compare leaching rates and volume-weighted mean concentrations, as well as maximum aboveground biomass among the crop treatments, including all years in each crop group. When a significant ($\alpha = 0.05$) difference was detected among the groups, we used the Tukey honest significant difference (HSD) post-hoc test to make pairwise comparisons among the groups. We lacked data on biomass in poplar and DON in grasses and poplar in some years, resulting in unequal sample sizes, and therefore we used the Tukey-Kramer method to make pairwise comparisons among the groups. Statements about correlations between variables refer to the Pearson product-moment correlation coefficient.

3. Results

3.1. Climate and hydrology

Air temperature recorded from the nearby KBS LTER weather station averaged 9.3 °C (mean for 2009–15), which was 0.2 °C warmer than the long-term average (1988–2015: 9.1 °C). Precipitation varied among crop-years (Fig. 1), with 2009 (725 mm) lower than the long-term average (1988–2015: 915 mm), while all of the other years (mean for 2010–15: 1000 mm) had higher precipitation than the long-term average.

Table 1
Nitrogen fertilizer applications to the cropping systems.

Crop-year	N applied (kg ha ⁻¹)					
	Corn	Switchgrass	Miscanthus	Native grasses	Poplar	Restored prairie
2009	158	0	77	0	0	0
2010	176	56	56	56	0	0
2011	172	56	76	56	157	0
2012	174	56	0	20	0	0
2013	173	57	57	57	0	0
2014	168	57	57	57	0	0
2015	172	57	57	57	0	0
Total	1193	339	380	303	157	0

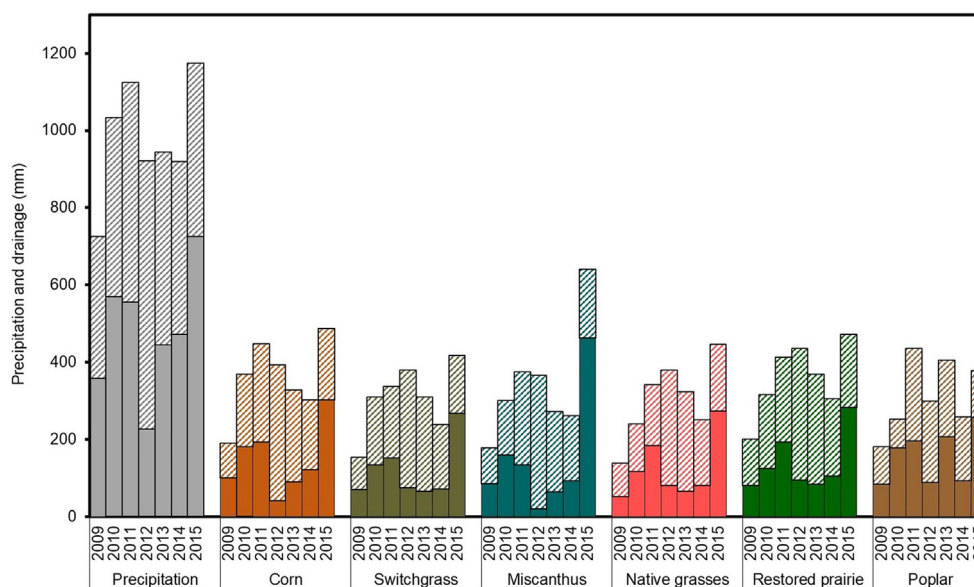


Fig. 1. Precipitation during each crop-year (gray bars) and total crop-year drainage (colored bars) of the different cropping systems. Precipitation and drainage for each crop-year are divided into the growing season (solid bar) and non-growing season (hatched bar). There were no significant differences in the drainage rates among treatments grouped across years (ANOVA $p = 0.85$).

Drainage rates simulated by SALUS over the seven years were not significantly different ($p = 0.85$) among cropping systems, with mean annual rates ranging from 315 mm (poplar) and 330 mm (perennial grasses) to 360 mm (corn) (Fig. 1, Table 2). Most drainage occurred in the non-growing season (~60%), except in 2015 when greater drainage occurred during the growing season as a result of particularly heavy rainfalls. Years with lower precipitation exhibited lower drainage (173 mm yr^{-1} in 2009) than years with higher precipitation (mean: 360 mm yr^{-1} for 2010–2015). Overall, modeled drainage rates for all cropping systems across years averaged 330 mm yr^{-1} , which is approximately 34% of the mean annual precipitation of 970 mm.

3.2. Growing season lengths and crop production

Growing season lengths for corn varied between 160 and 190 days. Except for poplar, the growing season lengths for the perennial cropping systems were more variable from year to year, with unusually early onset of growth in spring 2012 and relatively delayed onset in 2013 and 2014, although these differences were hardly visible in miscanthus (Fig. S2). Maximum aboveground biomass (Mg ha^{-1}) averaged across the seven years for the four replicate plots significantly differed ($p < 0.05$) among cropping treatments (Fig. 2). Poplar, miscanthus and corn were the most productive systems while switchgrass, native grasses and restored prairie were considerably less productive. The mean annual maximum aboveground biomass was lower in corn ($20.4 \pm \text{SE } 0.9 \text{ Mg ha}^{-1}$) than poplar ($30.8 \pm 1.9 \text{ Mg ha}^{-1}$) and miscanthus ($23.9 \pm 2.4 \text{ Mg ha}^{-1}$), but higher than the other perennial

systems (switchgrass: $7.7 \pm 0.9 \text{ Mg ha}^{-1}$; native grasses: $8.1 \pm 0.9 \text{ Mg ha}^{-1}$, and restored prairie: $6.8 \pm 0.8 \text{ Mg ha}^{-1}$). Interannual variability in maximum biomass of the perennial crops was high, with lower production in the rainfall-deficit years 2009 and 2012 in the perennial grass systems. Poplar, which was harvested during the 2013–14 winter, had an annual cumulative increment up until that time, after which a diversity of plants including coppicing poplar stems existed on the plot. Corn was markedly less productive in 2012 but not in 2009, when growing-season rainfall was not as low.

3.3. Leaching of dissolved nitrogen

Compared to NO_3^- and DON, NH_4^+ concentrations in our samples were low ($<0.07 \text{ mg N L}^{-1}$) and often undetectable ($<0.006 \text{ mg N L}^{-1}$) in the three crop-years (2013–2015) when measurements are available (Fig. S3), and therefore will not be discussed hereafter. Leaching rates and volume-weighted mean concentrations of DON and NO_3^- were influenced by N fertilization rates. Corn, which received the most fertilizer, leached more DON and NO_3^- (DON: $4.5 \pm \text{SE } 3.1$; NO_3^- : $35.3 \pm 11.5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$) than the moderately fertilized perennial grasses (DON and NO_3^- for switchgrass 0.8 ± 0.4 and 9.2 ± 1.3 , respectively; miscanthus 1.6 ± 0.8 and 7.6 ± 4.2 ; native grasses 1.9 ± 0.5 and $4.4 \pm 1.9 \text{ kg N ha}^{-1} \text{ yr}^{-1}$) and poplar (DON: 1.6 ± 0.5 ; NO_3^- : $7.2 \pm 2.1 \text{ kg N ha}^{-1} \text{ yr}^{-1}$). The unfertilized restored prairie leached the lowest NO_3^- ($2.4 \pm 1.7 \text{ kg N ha}^{-1} \text{ yr}^{-1}$) among perennial crops, while its DON leaching ($1.7 \pm 0.5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$) was similar to the other crops (Table 3, Fig. 3a). Across the seven crop-years, the volume-weighted mean concentrations of DON and NO_3^- in percolating water were higher in corn (DON: 1.0 ± 0.7 and NO_3^- : $9.5 \pm 2.9 \text{ mg N L}^{-1}$) than in perennial grasses and poplar (DON <1.0 and $\text{NO}_3^- < 3.3 \text{ mg N L}^{-1}$) (Table 3, Fig. 3b).

DON and NO_3^- leaching were influenced by time since establishment in the perennial cropping systems. NO_3^- leaching was higher in the initial two crop-years (means for 2009–2010 and 2010–2011: switchgrass $22.2 \pm \text{SE } 0.0$; miscanthus 17.4 ± 12.1 ; native grasses 7.9 ± 4.0 and poplar $14.6 \pm 0.0 \text{ kg N ha}^{-1} \text{ yr}^{-1}$) than in later years (means for 2011–15: switchgrass 4.0 ± 1.3 ; miscanthus 3.8 ± 1.0 ; native grasses 3.0 ± 1.1 and poplar $4.1 \pm 2.1 \text{ kg N ha}^{-1} \text{ yr}^{-1}$). In restored prairie, NO_3^- leaching was lower in the initial two years ($1.0 \pm 0.0 \text{ kg N ha}^{-1} \text{ yr}^{-1}$) than in later years ($3.0 \pm 1.8 \text{ kg N ha}^{-1} \text{ yr}^{-1}$) (Table 4, Fig. 3a).

Table 2

Mean annual drainage and percent of mean annual precipitation (970 mm) in each cropping system across the seven crop-years (2009–2015). There were no significant differences in drainage estimates among the cropping treatments ($p = 0.85$).

Cropping system	Mean drainage (mm yr^{-1})	% of mean annual precipitation
Corn	360	37
Switchgrass	310	32
Miscanthus	342	35
Native grasses	303	31
Restored prairie	358	37
Poplar	315	32
Grand mean	331	34

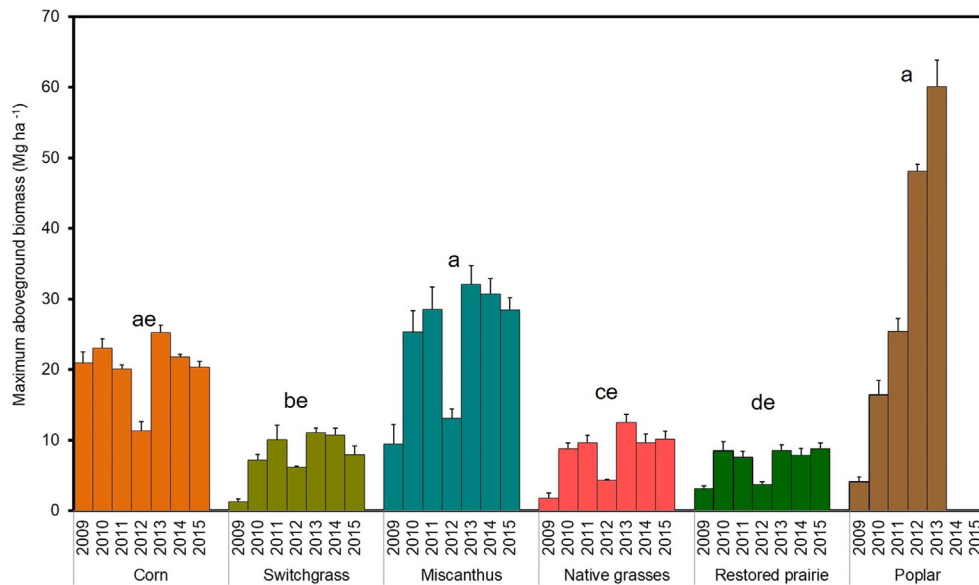


Fig. 2. Maximum aboveground biomass (often called aboveground net primary production) of the different cropping systems over the seven growing seasons. Corn biomass includes grain and stover. Poplar biomass was estimated from growth curves until the poplar was harvested in the winter of 2013–14. Error bars indicate the standard error for the four replicated blocks in each year. There were significant differences in the maximum biomass among treatments grouped across years (ANOVA $p < 0.05$). Where cropping systems share a similar letter, the means grouped across years are not significantly different as determined by the Tukey-Kramer post-hoc test ($\alpha = 0.05$).

These differences in NO_3^- leaching over time were driven both by concentrations and drainage rates (Table 5, Fig. 3b). In contrast to NO_3^- leaching, DON leaching did not show a consistent trend over time in the perennial cropping systems.

Depending on N fertilization rate and crop type, 14–38% of N added as fertilizer was lost to leaching (Fig. 4b), while the balance (unleached N) could either have accumulated in root biomass or soil organic matter, or been removed by biomass harvest, or lost to the atmosphere as gaseous N (Fig. 4a). The percentage of N leaching loss was highest in poplar (38% of added N) followed by corn (23%), while perennial grasses (excluding the unfertilized prairie) lost the lowest percentage (17%) (Fig. 4b). NO_3^- was the predominant form of leached N, comprising 59–92% of total N, with most of the balance lost as dissolved organic N and relatively little lost as NH_4^+ .

3.4. Leaching of dissolved organic carbon (DOC)

DOC leaching rates and volume-weighted mean concentrations over the seven crop-years varied significantly ($p < 0.05$) among the cropping systems. There was no consistent interannual trend except for lower DOC leaching and concentrations in the first year in all systems except prairie (Table 4, Fig. 5). DOC leaching rates and volume-weighted mean concentrations averaged $15.4 \text{ kg ha}^{-1} \text{ yr}^{-1}$ and 4.6 mg L^{-1} , respectively, over the seven crop-years. Poplar leached more DOC ($21.8 \text{ kg ha}^{-1} \text{ yr}^{-1}$ and 6.9 mg L^{-1}) than did the other cropping systems

(Table 3, Fig. 5). Unlike N leaching, DOC leaching was not directly related to N fertilization—leaching rates were similar in heavily fertilized corn ($13.9 \text{ kg ha}^{-1} \text{ yr}^{-1}$), moderately fertilized perennial systems ($16.9 \text{ kg ha}^{-1} \text{ yr}^{-1}$), and the unfertilized prairie ($11.1 \text{ kg ha}^{-1} \text{ yr}^{-1}$).

Across all cropping systems, we observed positive but rather weak relationships between DOC and DON concentrations ($r = 0.26$, $p > 0.05$), as well as between DON and NO_3^- concentrations ($r = 0.47$, $p < 0.05$, Fig. S4). DOC concentrations were not correlated with NO_3^- concentrations (data now shown).

4. Discussion

Our results show that perennial cropping systems grown in close proximity on a well-drained sandy loam soil, moderately fertilized with N and harvested annually, leach less N than the more heavily fertilized annual corn system. In both corn and the perennial systems, NO_3^- was the predominant form of leached N. Leaching of DON was markedly higher in the corn system, albeit still small relative to NO_3^- leaching (Fig. 4). In contrast, DOC leaching was relatively unaffected by crop type or N fertilization, except that it was higher in the poplar system (Table 3, Fig. 5).

Our study builds on earlier reports of NO_3^- leaching from annual corn-soybean-wheat rotations (Syswerda et al., 2012) and perennial bioenergy cropping systems at this study site (Hussain et al., 2019), but is distinctive because we report measurements of leaching of

Table 3

Mean leaching rates and volume-weighted mean concentration of NO_3^- , DON and DOC in different cropping systems over the seven crop-years (2009–2015). Where cropping combinations share a similar letter, the means grouped across years are not significantly different as determined by the Tukey honest significant difference post-hoc test ($\alpha = 0.05$). When some years of data were missing, the Tukey-Kramer method was used to make pairwise comparisons among the groups. Standard errors represent the mean of four replicate plots. In the 2009 and 2010 crop-years, data from some replicates are missing ($n = 1$ or 2).

Cropping system	Leaching rate ($\text{kg ha}^{-1} \text{ yr}^{-1}$)			Volume-wtd mean concentration (mg L^{-1})		
	NO_3^- -N	DON	DOC	NO_3^- -N	DON	DOC
Corn	35.3 ± 11.5^a	$4.5 \pm 3.1^{\text{ns}}$	$13.9 \pm 3.5^{\text{ab}}$	9.4 ± 2.9^a	$1.24 \pm 0.7^{\text{ns}}$	$3.80 \pm 1.1^{\text{abcd}}$
Switchgrass	9.2 ± 1.3^b	$0.8 \pm 0.4^{\text{ns}}$	9.5 ± 2.3^a	3.3 ± 0.4^b	$0.26 \pm 0.1^{\text{ns}}$	$2.98 \pm 0.7^{\text{baf}}$
Miscanthus	7.6 ± 4.2^b	$1.6 \pm 0.8^{\text{ns}}$	$18.1 \pm 6.3^{\text{ab}}$	3.1 ± 1.8^b	$0.53 \pm 0.2^{\text{ns}}$	$5.13 \pm 1.6^{\text{cde}}$
Native grasses	4.4 ± 1.9^b	$1.9 \pm 0.5^{\text{ns}}$	$18.4 \pm 6.0^{\text{ab}}$	2.3 ± 1.1^b	$0.55 \pm 0.2^{\text{ns}}$	$5.79 \pm 1.6^{\text{dc}}$
Poplar	7.2 ± 2.1^b	$1.6 \pm 0.5^{\text{ns}}$	21.8 ± 6.3^b	3.0 ± 0.6^b	$0.64 \pm 0.3^{\text{ns}}$	6.91 ± 1.9^e
Restored prairie	2.4 ± 1.7^b	$1.7 \pm 1.0^{\text{ns}}$	$11.1 \pm 3.0^{\text{ab}}$	0.6 ± 0.4^b	$0.47 \pm 0.1^{\text{ns}}$	$3.13 \pm 0.8^{\text{fab}}$

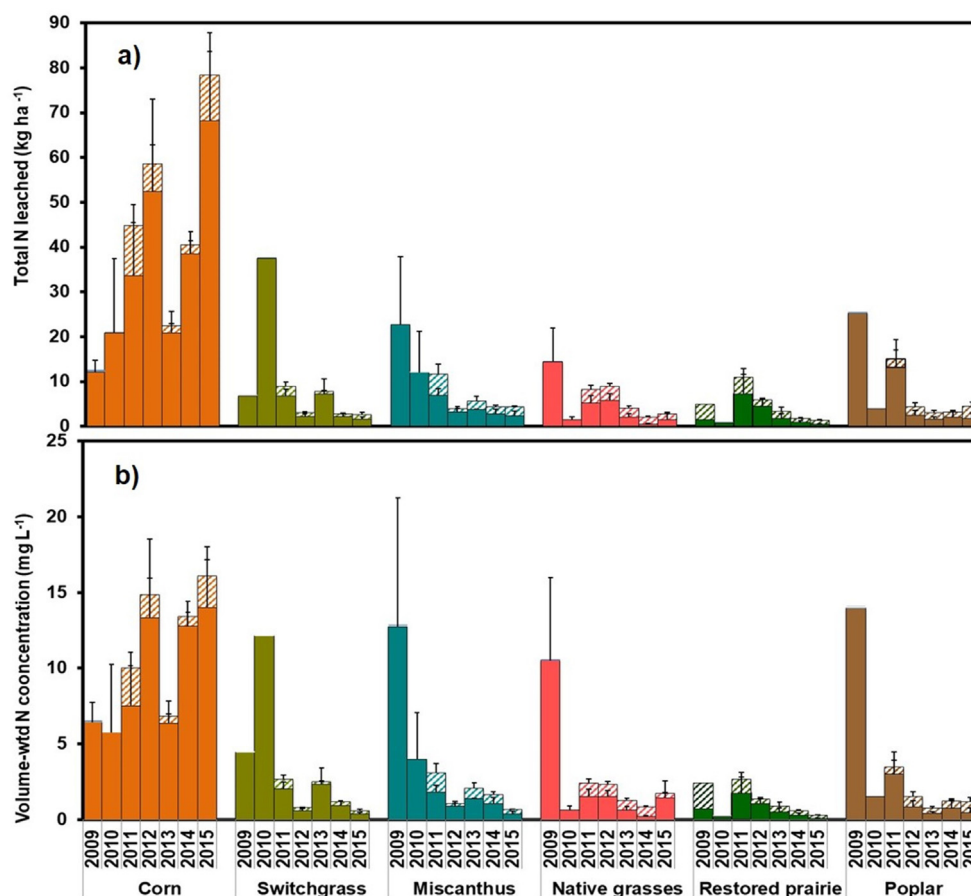


Fig. 3. Dissolved N leaching rate (a) and volume-weighted mean N concentration (b) in the leachate of different cropping systems. Hatched bars indicate dissolved organic nitrogen (DON) and solid bars indicates nitrate (NO_3^-) concentrations. Ammonium (NH_4^+) concentrations were much lower and often undetectable ($<0.07 \text{ mg N L}^{-1}$) (Fig. S3). Bars show the standard error of the means of four replicate plots. In 2009 and 2010 crop-years, data from some replicates are missing ($n = 1$ or 2). Years on x-axis refer to crop-years. Statistical comparisons of the leaching rates and volume-weighted mean concentrations among treatments are given in Table 3.

NH_4^+ , DOC and DON as well as NO_3^- . Our estimate of NO_3^- leaching from the no-till continuous corn ($35 \text{ kg ha}^{-1} \text{ yr}^{-1}$) is similar to the mean annual rate reported by Syswerda et al. (2012) for no-till corn-soybean-wheat rotations on a nearby site ($41 \text{ kg ha}^{-1} \text{ yr}^{-1}$).

Dissolved organic compounds leached from plant litter and soils are likely to be heterogeneous in composition and to include compounds spanning a gradient of lability to microbial decomposition, and the source of the biomass might be expected to result in compositional differences in leached dissolved organic matter (Jones and Kielland, 2002). Nevertheless, our results showed similar rates of total DOC leaching among the cropping systems, except for higher rates from poplar. For all crops the first crop-year had lower rates, which could reflect the lower initial production of the crops (Table 4), but that year also has less available DOC data, with only one or two replicate plots. The

similarity in DOC leaching rates between the annual corn and the perennial herbaceous systems may be due to the annual harvest of most aboveground biomass. As has been shown in other studies (Bonin and Lal, 2014; Fortier et al., 2015; Chimento et al., 2016), poplar may have leached more DOC because of the high biomass accrual over time until its one-time harvest, as well as annual litterfall from the poplar trees and the plants that replaced it after the trees were harvested and before the new stand shaded the understory.

In forest and grassland ecosystems DOC leaching losses can be a significant term in the carbon budget (Fahey et al., 2005; Chapin et al., 2006), and a few studies in agricultural systems also indicate significant losses (Kindler et al., 2011; Manninen et al., 2018), but that does not appear to be the case in these cropping systems. DOC leaching losses averaged $14.9 \text{ kg C ha}^{-1}$ (1.49 g C m^{-2}) across all cropping systems. If not

Table 4
Mean leaching rates of NO_3^- , DON and DOC during the initial two crop-years (2009 and 2010) since perennial system establishment compared to later crop-years (2011–2015). Standard error represents the mean of four replicate plots, except in the 2009 and 2010 crop-years, when data from some replicates are missing ($n = 1$ or 2).

Cropping systems	Leaching ($\text{kg ha}^{-1} \text{ yr}^{-1}$) during 2009 and 2010 crop-years			Leaching ($\text{kg ha}^{-1} \text{ yr}^{-1}$) during 2011–2015 crop-years		
	$\text{NO}_3^- \text{-N}$	DON	DOC	$\text{NO}_3^- \text{-N}$	DON	DOC
Corn	16.5 ± 9.5	0.19	13.0 ± 1.0	42.7 ± 12.3	6.1 ± 3.1	14.3 ± 4.5
Switchgrass	22.2	0.07	9.0	4.0 ± 1.3	1.0 ± 0.4	9.7 ± 2.3
Miscanthus	17.4 ± 12.1	0.11	11.1	3.8 ± 1.0	2.1 ± 0.8	20.1 ± 6.3
Native grasses	7.9 ± 4.0	0.11	10.2	3.0 ± 1.0	2.2 ± 0.5	21.6 ± 6.0
Restored prairie	1.0	1.74	7.3	3.0 ± 1.8	1.7 ± 0.5	12.6 ± 3.0
Poplar	14.6	0.30	14.9	4.1 ± 2.0	1.9 ± 1.0	24.6 ± 6.3
Mean of perennial grasses	12.6	0.50	9.4	3.5	1.8	16

Table 5

Volume-weighted mean concentrations of NO_3^- , DON and DOC during the initial two crop-years (2009 and 2010) since perennial system establishment compared to later crop-years (2011–2015). Standard error represents the mean of four replicate plots. In 2009 and 2010 crop-years, data from some replicates are missing ($n = 1$ or 2).

Cropping systems	Concentration (mg L^{-1}) during 2009 and 2010 crop-years			Concentration (mg L^{-1}) during 2011–2015 crop-years		
	NO_3^- -N	DON	DOC	NO_3^- -N	DON	DOC
Corn	6.1 ± 2.9	0.1	4.0 ± 0.4	10.8 ± 3.0	1.4 ± 0.7	3.7 ± 1.1
Switchgrass	8.2	0.04	3.3	1.2 ± 0.4	0.3 ± 0.1	2.8 ± 0.7
Miscanthus	8.4 ± 5.8	0.06	4.3	1.1 ± 0.2	0.6 ± 0.2	5.5 ± 1.6
Native grasses	5.6 ± 2.9	0.08	5.0	1.0 ± 0.4	0.7 ± 0.1	6.0 ± 1.6
Restored prairie	0.4	0.87	3.0	0.7 ± 0.4	0.4 ± 0.1	3.2 ± 0.8
Poplar	7.8	0.17	6.6	1.1 ± 0.5	0.5 ± 0.2	7.0 ± 1.9
Mean of perennial grasses	5.7	0.3	3.9	1.0	0.5	4.4

accounted for, DOC leaching would appear as net C sequestration in ecosystem carbon budgets. Based on eddy covariance methods, [Abraha et al. \(2018\)](#) reported cumulative net ecosystem exchange (NEE) rates of carbon dioxide over seven years for similar corn, switchgrass, and prairie fields near our study site, accounting for annual harvest of most aboveground biomass. Results depended on prior land use; in the case of switchgrass and prairie planted on former cropland, net CO_2 uptake (not including harvest) was observed with 8-year cumulative values of -478 ± 101 and $-735 \pm 88 \text{ g C m}^{-2}$, respectively. Our mean DOC leaching rate would amount to just $\sim 2\%$ (-11.9 g C m^{-2}) of the NEE observed in those fields over eight years.

Although NO_3^- represented the majority of leached N in all cropping systems, accounting for 59–92% of total N leaching, DON was a significant component as well ([Fig. 4a](#)). Corn showed higher DON leaching in some years that could be related to the higher N fertilization ([Fig. 3](#)), as has been found in other studies ([Jiao et al., 2004](#); [van](#)

[Kessel et al., 2009](#); [Salazar et al., 2019](#)). DON concentrations were not strongly correlated with DOC concentrations, suggesting significant compositional variability in the dissolved organic matter.

Leached DON is normally less available for plants and microbes than dissolved inorganic forms, as has been shown in agroecosystems ([He et al., 2017](#); [Salazar et al., 2019](#)) as well as unmanaged ecosystems ([Jones et al., 2004](#); [McDowell et al., 2004](#); [Dijkstra et al., 2007](#)). Our observations of significant concentrations of NO_3^- in leachate suggest that plant and microbial N demands could have been met without utilization of DON.

The overall mean annual drainage rates predicted for these cropping systems by the SALUS model over the seven crop-years ($303\text{--}360 \text{ mm yr}^{-1}$, or 31–37% of mean annual precipitation; [Table 2](#)) are in agreement with previous studies of annual and perennial crop evapotranspiration estimated by soil water monitoring at the same site ([Hamilton et al., 2015](#)), and from eddy covariance analyses of

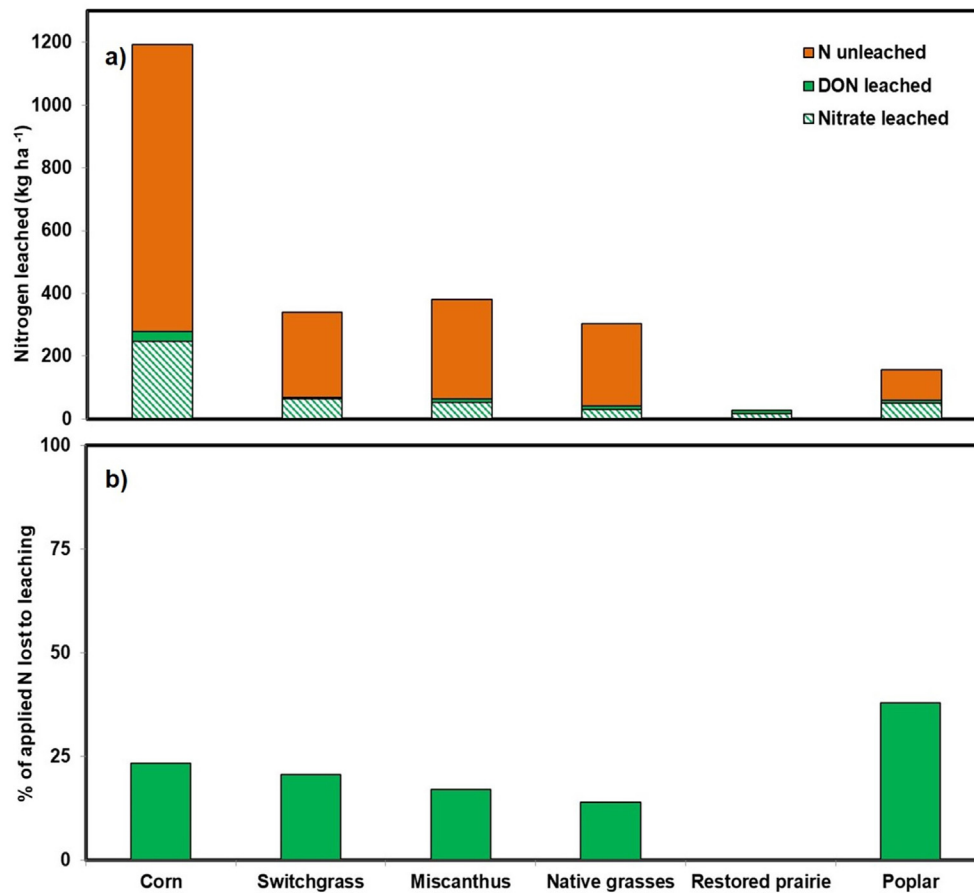


Fig. 4. Summary of total amount and forms of N leached over the seven years (a) and the % of applied N nitrogen lost to leaching (b) over the seven years for the different cropping systems. The fate of unleached N potentially includes removal in harvest, accumulation in root biomass and soil organic matter, and gaseous emissions.

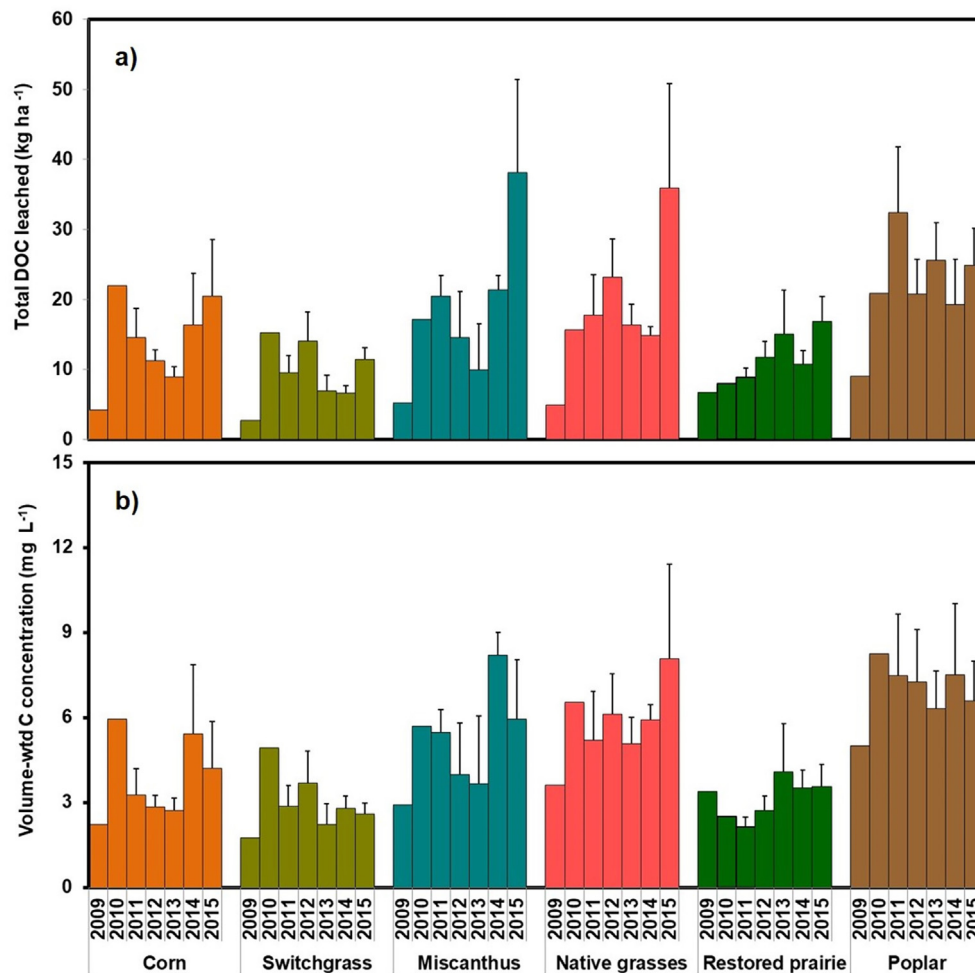


Fig. 5. Dissolved organic carbon (DOC) leaching rate (a) and volume-weighted mean concentration (b) in the leachate of different cropping systems. Bars show the standard error of the means of four replicate plots. Water samples were not available from all replicates, resulting in no error bars in some years. Statistical comparisons of the leaching rates and volume-weighted mean concentrations among treatments are given in Table 3.

water vapor fluxes for nearby plantings of corn, switchgrass, and prairie on similar soils (Abraha et al., 2015). These conclusions are also supported by the observation that watershed water balances in the vicinity have not changed detectably in spite of historical land cover changes (Hamilton et al., 2018).

Variation in total N leaching rates was largely driven by differences in NO_3^- leaching, which in turn were directly related to N fertilization, being highest in corn (Fig. 3a). This pattern has been reported in many other studies (e.g., Carefoot and Whalen, 2003; Jiao et al., 2004; van Kessel et al., 2009; Manninen et al., 2018; Salazar et al., 2019). With the exception of the prairie, which was not fertilized, NO_3^- leaching was highest in the perennial systems in the first 1–2 years after crop establishment (Table 4, Fig. 3a), perhaps reflecting less efficient use of the available N from added fertilizer and soil organic matter mineralization because the crops had not yet reached their potential aboveground (Fig. 2; Sanford et al., 2016) and belowground biomass (Sprunger et al., 2017). Higher NO_3^- leaching in the initial crop establishment phase has been observed in other studies of perennial grass crops (McIsaac et al., 2010; Smith et al., 2013; Diaz-Pines et al., 2016). Concentrations of NH_4^+ in the soil solutions were low and often below our detection limit, and therefore leaching of NH_4^+ was a very small component of total N leaching compared to NO_3^- and DON (Fig. S3).

Averaged over the seven crop-years, the fertilized perennial grass crops leached more N than the unfertilized prairie, primarily in the form of NO_3^- (Table 3, Figs. 3, 4a). Poplar, which was fertilized only once but heavily, lost the greatest percentage of added N to leaching.

Poplar plantations may not always leach as much N as we observed because they are not always fertilized (Nelson et al. 2019), or they may be fertilized at lesser amounts over multiple years (Stanturf et al., 2001; Coleman et al., 2006), but comparative studies of leaching are lacking.

In conclusion, the annual and perennial cropping systems leached similar amounts of DOC (with the exception of poplar), whereas DON leaching was markedly higher in the more heavily fertilized corn system, which also leached the most NO_3^- . Nitrate comprised the majority of total N leaching in all systems, and the perennial crops leached more NO_3^- in the first few years after planting. Requiring only one third or less of the N fertilization compared to corn, perennial bioenergy crops can reduce N leaching and consequent movement into aquifers and surface waters (Robertson et al., 2017; Schulte et al., 2017). While improved N management practices such as cover crops (Walmsley et al., 2018), use of slow-release N fertilizer (Noellsch et al., 2009), and vegetated buffer strips (Sahu and Gu, 2009) can also minimize nutrient losses from agroecosystems, perennial bioenergy crops could be part of a portfolio of options and may be particularly well suited to excessively permeable soils that are prone to N leaching.

CRediT authorship contribution statement

Mir Zaman Hussain: Investigation, Formal analysis, Writing - original draft. **G. Philip Robertson:** Conceptualization, Project administration, Writing - review & editing. **Bruno Basso:** Methodology. **Stephen K.**

Hamilton: Conceptualization, Methodology, Supervision, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2020.139379>.

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Further Reading

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