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Dissolved Organic Nitrogen: An Overlooked Pathway of Nitrogen Loss from Agricultural Systems?

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Conventional wisdom postulates that leaching losses of N from agriculture systems are dominated by NO_3^-. Although the export of dissolved organic nitrogen (DON) into the groundwater has been recognized for more than 100 yr, it is often ignored when total N budgets are constructed. Leaching of DON into stream and drinking water reservoirs leads to eutrophication and acidification, and can pose a potential risk to human health. The main objective of this review was to determine whether DON losses from agricultural systems are significant, and to what extent they pose a risk to human health and the environment. Dissolved organic N losses across agricultural systems varied widely with minimum losses of 0.3 kg DON $ha^{-1}yr^{-1}$ in a pasture to a maximum loss of 127 kg DON $ha^{-1}yr^{-1}$ in a grassland following the application of urine. The mean and median values for DON leaching losses were found to be 12.7 and 4.0 kg N $ha^{-1}yr^{-1}$, respectively. On average, DON losses accounted for 26% of the total soluble N (NO_3^- plus DON) losses, with a median value of 19%. With a few exceptions, DON concentrations exceeded the criteria recommendations for drinking water quality. The extent of DON losses increased with increasing precipitation/irrigation, higher total inputs of N, and increasing sand content. It is concluded that DON leaching can be an important N loss pathway from agricultural systems. Models used to simulate and predict N losses from agricultural systems should include DON losses.

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In the second half of the 19th century, before the discovery of biological nitrogen fixation in 1888, detailed total N budget studies were performed to determine the source of N in agricultural systems (Burris, 1974). Lawes and coworkers (1881) conducted a remarkably thorough study on the amounts of various N compounds in rain and drainage waters at the Rothamsted Station in the United Kingdom (see also Murphy et al., 2000; Burris, 1974). Drainage water, collected at three different depths, was analyzed for NO_3^-, NO_2^-, NH_4^+, and N present in the dissolved organic matter. Drainage water collected at 150 cm depth showed a total N content of 21.03 mg L^{-1}, of which 20.40 mg L^{-1} was in the form of NO_3^- plus NO_2^-, 0.08 mg L^{-1} as NH_4^+, and 0.55 mg L^{-1} as organic N. The dissolved organic matter had a C to N ratio of 3.1 and this ratio was shown to increase with increasing depth of drainage water collection. Total N leaching losses were calculated to be 50 kg N ha^{-1} yr^{-1}. The authors considered the leaching losses of DON to be small, but noticed that losses increased when turbidity increased and that in all cases the dissolved organic matter was highly nitrogenous. Lawes et al. (1881) also alluded to the possible role of dissolved organic N in plant nutrition but concluded that "very little was known at present."

While leaching losses of DON from agricultural fields have now been recognized for more than 125 yr, most N loss studies in agricultural systems have not measured DON as a potential pathway of N loss. Furthermore, many soil N cycling and leaching simulation models used for agricultural systems do not contain a subcomponent for simulating the leaching of organic N compounds (Korsaeth et al., 2003). Similarly, when nutrient budgets for pastures or cropping systems are constructed, losses of DON are not considered (Ghani et al., 2007). This is in sharp contrast with many nonagricultural systems such as forests, where DON has been considered to be a major component of the N cycle for many years. Numerous studies conducted in forested ecosystems have shown that DON losses can be substantial (Campbell et al., 2000; Neff et al., 2000, 2002; Qualls et al., 2000; Perakis and Hedin, 2002). In many instances DON losses from forested ecosystems, and in particular for undisturbed forest systems, were found to exceed NO_3^- leaching losses (Hedin et al., 1995; Qualls et al., 2000; Pe-

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Abbreviations: DON, dissolved organic nitrogen; EU, European Union; HMW, high molecular weight; LMW, low molecular weight; SON, soluble organic nitrogen.

rakis and Hedin, 2002; Lajtha et al., 2005). Neff et al. (2003) postulated that "over centuries, DON leaching may represent a significant 'leak' of N as plant and microbes cannot prevent DON losses, even in times of high N demand." Nitrogen is considered to have leaked out of the system when the biological system cannot fully prevent the loss of N and therefore leakage cannot be avoided. Leakage of N is an integral part of the global N cycle. However, N is considered to be lost when the loss of N can be controlled potentially by biological demand (Neff et al., 2003). In this case, the loss of this N does not have to be considered as an integral part of the global N cycle and can be avoided. For example, since NO_3^- can be taken up by plants, losses of NO_3^- via leaching would be considered a loss. Whereas it is clear from research conducted in forest ecosystems that DON is an integral and important part of the N cycle and can be the major loss pathway for N, the significance of DON in agricultural production systems as a potential pathway for losses remains much less understood and has only received scant attention (Murphy et al., 2000).

The impact of DON losses from agricultural fields on water quality has already been shown for the Chesapeake Bay area where the concentrations of DON were related to the surrounding area of agricultural land (Jordan et al., 1997).

One possible explanation for the apparently limited attention given to leaching losses of DON in agriculture may be driven by the common understanding that NO_3^- is the predominant form of plant available N in agricultural soils. Fertilizer N, whether applied in the oxidized or reduced form, will ultimately be present in the soil as NO_3^-. Similarly, when organic N amendments are applied, the organic N will largely be converted into NO_3^-, following mineralization and subsequent nitrification, making NO_3^- the dominant form of soluble N in agricultural systems. Because of its high solubility and the dominant form of soluble N in the soil, it has become the conventional wisdom that most N leaching losses will also occur as NO_3^-.

Terminology and Background

A distinction has to be made between soluble organic nitrogen (SON) and DON (Murphy et al., 2000). Soluble organic N is soil N that is extracted from the soil using water, KCl, electroultrafiltration (EUF), K_2SO_4, $CaCl_2$, or any other extractants. Dissolved organic N is defined as the fraction of SON fraction which is collected in situ using a lysimeter or suction cup among other devices, and where no extractant is used. In the literature, however, the terms SON and DON, as defined here, are used interchangeably; often the term DON is used when soluble soil organic N was obtained with an extractant. As Murphy et al. (2000) pointed out, the chemical composition of these two soluble soil N pools, DON and SON, are not similar and may differ in both quantitative and qualitative aspects. Soluble organic N is in equilibrium with organic N adsorbed on clay colloids, thus the pH of the extraction, as well as its ionic strength and composition, will affect adsorption/desorption and the equilibrium and thereby the concentration of SON in solution (Haynes, 2005). Using different extractants yields different soluble N pools and the relationship between SON extracted by water or using 2 mol L^{-1} KCl remains unclear (Haynes, 2005). Both pools, DON and SON, are at least partially composed of easily decomposable, mineralizable N and have a significant effect on the size of the inorganic soil N pools: NH_4^+ and NO_3^- (Mengel et al., 1999). When K_2SO_4 is used as the extractant, soluble organic C and N can also provide an estimate of C and N in the microbial biomass (Vance et al., 1987).

Of the total amount of N present in the agricultural soil, 0.15 to 0.19% is in the form of DON (Haynes, 2000). Soluble soil organic N cannot be measured directly by extraction but is calculated by subtracting the inorganic N pool from the total soluble N pool. The classical and cumbersome Kjeldahl digestion can be used to determine total soluble N. However, more often the persulfate ($K_2S_2O_8$) oxidation method is used which converts both inorganic and organic N to NO_3^- followed by a colorimetric analysis (Smart et al., 1981; Cabrera and Beare, 1993). Soluble organic N is then calculated as the difference between total soluble N and dissolved inorganic N (NO_3^-, NO_2^-, NH_4^+).

Dissolved organic N can also be separated into hydrophobic and hydrophilic compounds. The hydrophilic fraction of DON extracted from a soil planted with cabbage (*Brassica oleracea*) was the dominant fraction, 78%, of organically bound N (Moller et al., 2005). Largely similar hydrophilic fractions were also found in a primary and secondary forest and at a site that had undergone reforestation (Moller et al., 2005). When different rates of debris were added to a forest floor, the concentrations of leached hydrophilic DON and DOC were related ($R^2 = 0.86$), whereas the concentrations of hydrophobic DON and DOC were not related (Lajtha et al., 2005). Differential C input via roots and litter in terms of quantity and chemical composition by depth via roots and litter may lead to these differences in hydrophobic and hydrophilic DON and DOC relationships. The DOC-to-DON ratio in soil solutions, collected by centrifugation at high speed and obtained from surface soils of 70 sites under different vegetation and fertilizer management practices, was found to be 16 ± 4, and was similar for all land uses with the exception of forest which showed a higher ratio (Christou et al., 2005).

Jones et al. (2004) hypothesized that there are two distinct DON pools in the soil. The low molecular weight (LMW)-DON pool which is composed of mainly free amino acids and proteins, has a high turn-over rate and does not accumulate in the soil. This pool is directly related to ammonification and nitrification activities as it serves as a substrate for these processes. The high molecular weight (HMW)-DON pool is rich in humic substances, has a slower turn-over rate, and is the predominant source of DON in groundwater and streams.

Collection of Dissolved Organic Nitrogen

A recent review listed sampling devices generally used to collect in situ extraction of soil water and hence DON (Weihermuller et al., 2007). Six different sampling devices were discussed: porous cups, porous plates, capillary wicks, pan lysimeters, resin boxes, and lysimeters. For each device, specific advantages and disadvantages were listed. No recommendation for a single best approach could be made. In short, the main disadvantage of the suction cup is that it remains unknown how well the solution in the suction cup represents the soil solution. Installation of the suction cup might affect the natural water flow and generally leads to a bias toward larger soil pore water being sampled at the expense of smaller pores. Also, suction cups have been shown to change the composition and the quantity of the organic material because of sorption (Hansen and Harris, 1975; Raulund-Rasumusses, 1989). However, various precautions can be taken to minimize sorption and contamination in suction cups (Weihermuller et al., 2007).

Lysimeters are devices that contain disturbed or undisturbed soil columns placed in confined containers. Sizes of lysimeters can vary widely, ranging from 10 cm diam. to large containers holding many tonnes of soil that are placed on large weighing scales with below ground access (Di et al., 1998). Large controlled drainage plots encompass small field plots where the soil is no longer contained in an enclosure but where the borders are lined with plastic (Saarijarvi et al., 2004). A major drawback of lysimeters, when used to measure the total amount of DON leached, is that DON originating from lateral water and solute fluxes is excluded. Moreover, the vertical boundaries can create edge flow effects and preferential flow paths. Weihermuller et al. (2007) concluded that "it seems difficult and perhaps impossible to obtain pore water samples which are not altered or biased by the sampling process". Lysimeters, as well as suction cups, can lead to underestimations of the actual leachate-N losses because of the general absence in a lysimeter of by-pass or preferential flow. For example, earthworms like *Lumbricus terrestris* and other deep-burrowing, anecic earthworms species form vertical semi-permanent burrows which can cause preferential flow or macropore flow of water and nutrients. Their exclusion from the soil in lysimeters might lead to a reduction in solute transport (Li and Ghodrati, 1995; Lachnicht et al., 1997).

To avoid potential problems associated with the scaling up of results obtained from a relatively small experimental unit to a larger area, such as a field or farm, leachates need to be collected from larger sampling areas. Installing a drainage system and collecting drainage water from agricultural fields is a more reliable method for obtaining accurate data on the extent of DON fluxes. However, its main drawbacks are the operational and financial resources needed for such a setup (Murphy et al., 2000). Once the drainage is installed, the subsurface tiles need constant attention and have to be maintained (Cannell et al., 1984). Fluxes of N losses are determined by measuring the flow rate, preferably automatically, at regular intervals and analyzing water for DON concentrations (Lawes et al., 1881; Kanwar et al., 1999; Randall and Vetsch, 2005). The tile system, however, can also lead to an underestimation of the total DON losses if the tiles only intercept a portion of the drainage water. Therefore, a further scaling up can be done by sampling for DON at the catchment scale. By sampling the inflow and outflow waters of a confined catchment, the impact of a particular land use on the amount of DON exported from the catchment can be measured. Watson et al. (2000) measured total DON (called SON by the authors) concentrations in small rivers which drained six different catchment areas, encompassing a total area of 4453 km^2, predominately present in grassland. The advantage of sampling at the catchment level is that it provides a realistic estimate of DON losses from agricultural fields. The main disadvantage is that the system is difficult to manipulate, for example to determine the effect of a particular management practice on DON losses. As farming practices in an entire watershed area will likely not be uniform, it remains difficult to identify the source of DON. Moreover, in-stream processes can also change the DON concentrations in the river water and will make it more difficult to determine the impact of agricultural management practices on DON losses.

Leaching of Dissolved Organic Nitrogen

A literature survey of peer-reviewed publications on DON losses measured under field conditions in agricultural systems was performed using the ISI-Web of Science research database. We would like to make clear here that losses of DON from forest ecosystems or other nonagricultural systems were not included in the survey. Losses of DON from nonagricultural systems have been reported and reviewed extensively elsewhere (Sollins et al., 1980; Sollins and McCorison, 1981; Qualls and Haines, 1991; Qualls et al., 1991, 2000; Hedin et al., 1995; Neff et al., 2002, 2003; Perakis and Hedin, 2002; Cooper et al., 2007). In this review, only field studies of agricultural systems that reported annual DON losses per hectare or reported data that made it possible to calculate annual DON losses were included. Although the study by Lawes et al. (1881) is not included in the ISI-Web of Science research database, the results of this seminal study have been included here. A total 16 studies were found which reported annual DON as well as NO_3^- losses per hectare (Table 1). It is of interest to note that with the exception of the study of Lawes et al. (1881), all other studies were published in 2000 or thereafter. It is possible that we overlooked some peer reviewed research findings on DON losses from agricultural systems that were published in the 20th century. However, our search results indicate that very limited attention was paid to DON losses from agricultural systems in the previous century. Murphy et al. (2000) among others also concluded that very little is known on the role DON in the N cycle in agricultural soils, especially as compared to (semi-)natural systems like forested ecosystems.

All but 3 of the 16 studies used suction cups or lysimeters to collect leachates. The remaining three studies measured DON losses using a tile drain system and at the catchment scale. This is somewhat in contrast to studies conducted in forest ecosystems on DON losses where measurements are mostly taken at the catchment level (Sollins et al., 1980; Edwards et al., 2000; Perakis and Hedin, 2002). Lack of catchments under a single, uniform agricultural management practice, in addition to practical difficulties when superimposing treatments across catchments areas are likely to be the main reasons why leachates for agricultural systems are often collected with lysimeters or suction cups.

Observed losses of DON from agricultural systems were highly variable and ranged from 0.3 kg N $ha^{-1}yr^{-1}$ in a grass clover system (Saarijarvi et al., 2004) to a maximum of 127 kg N ha^{-1} yr^{-1} in a pasture following the application of urine (Wachendorf et al., 2005; Table 1). When averaged across all experimental sites and treatments, the mean value for DON losses was 12.7 kg N ha^{-1} yr^{-1}, with a median value of 4.0 kg N ha^{-1} yr^{-1}. When the DON leaching study applying high rates of manure and urine, mimicking manure and urine patches in the field was

Table 1. Losses of dissolved organic nitrogen (DON), nitrate, and dissolved organic carbon (DOC) across a diverse array of agricultural systems with various rates of N inputs.

Cropping system	Method	Precipitation	Texture	N input		Leached				Reference
				Manure	Inorganic	DON	Nitrate	DOC	DON	
		mm yr^{-1}		kg ha^{-1} yr^{-1}					%†	
Ryegrass/maize‡	Suction cup	742	Sandy	108	62	8.6	50	30§	15	Siemens et al., 2002, 2003
Ryegrass/maize‡	Suction cup	742	Sandy	161	41	9.2	59	38	13	Siemens et al., 2002, 2003
Fallow‡	Suction cup	742	Sandy	0	0	4.7	2.5	33	65	Siemens et al., 2002, 2003
Fallow/sheep‡	Suction cup	742	Sandy	0	0	4.0	17	96	19	Siemens et al., 2002, 2003
Maize/soybean/chisel§	Lysimeter	900	Silt-loam	–	150	0.4	0.8	nr#	33	Shuster et al., 2003
Maize/soybean/chisel¶	Lysimeter	900	Silt-loam	–	150	3.8	6.4	nr	37	Shuster et al., 2003
Maize/soybean/ridge§	Lysimeter	900	Silt-loam	–	150	2.1	4.4	nr	32	Shuster et al., 2003
Maize/soybean/ridge¶	Lysimeter	900	Silt-loam	–	150	3.1	8.9	nr	26	Shuster et al., 2003
Cabbage	Suction cup	1400	Silt-clay-loam	–	40–60	0.9	4.1	17.2	18	Moller et al., 2005
Grass-clover/fallow	Suction cup	nr	Coarse sandy	120	–	31	316	216	9	Vinther et al., 2006
Grass-clover	Suction cup	nr	Sandy loam	0	0	3.3#	10.5	28.3	24	Vinther et al., 2006
Grass-clover/barley	Suction cup	nr	Coarse sandy	120	–	20	303	174	6	Vinther et al., 2006
Grass-clover	Lysimeter	627	Sandy loam	–	220	0.3	0.9	nr	25	Saarijarvi et al., 2004
Grass-clover/roundup	Lysimeter	627	Sandy loam	–	–	2.8	26.4	nr	10	Saarijarvi et al., 2004
Grass/grass clover	Lysimeter	567††	Sandy loam	65	110	0.5	12.3	nr	4	Saarijarvi et al., 2007
Plowed-barley/grass	Lysimeter	545††	Sandy loam	–	–	5.5	40.2	nr	12	Saarijarvi et al., 2007
Grassland-mono‡‡	Lysimeters	660	nr	–	0	1.6	18.8	nr	8	Dijkstra et al., 2007
Grassland-mono‡‡	Lysimeter	660	nr	–	40	3.0	30.0	nr	9	Dijkstra et al., 2007
Grassland-diverse‡‡	Lysimeter	660	nr	–	0	3.5	2.3	nr	60	Dijkstra et al., 2007
Grassland-diverse‡‡	Lysimeter	660	nr	–	40	3.8	2.8	nr	58	Dijkstra et al., 2007
Pasture	Basin	nr	Sandy-clay-loam	–	100	4	13	nr	24	Watson et al., 2000
Pasture	Basin	nr	Sandy-clay-loam	–	200	4.3	21.5	nr	17	Watson et al., 2000
Pasture	Basin	nr	Sandy-clay-loam	–	300	4.3	35	nr	11	Watson et al., 2000
Pasture	Basin	nr	Sandy-clay-loam	–	400	4.5	57.6	nr	7	Watson et al., 2000
Pasture	Basin	nr	Sandy-clay-loam	–	500	3.5	63	nr	5	Watson et al., 2000
Pasture§§	Lysimeter	824	nr	1030¶¶	–	127	542	nr	19	Wachendorf et al., 2005
Pasture§§	Lysimeter	824	nr	1052##	–	23	94	nr	20	Wachendorf et al., 2005
Pasture	Lysimeter	1200	Silty sand	–	200	2.2	7.7	nr	22	Korsaeth et al., 2003
Pasture	Lysimeter	1200	Silty sand	195	–	4.8	15.7	nr	23	Korsaeth et al., 2003
Pasture	Lysimeter	1200	Silty sand	127	80	2.5	11.6	nr	18	Korsaeth et al., 2003
Pasture	Lysimeter	1200	Coarse sand	127	80	13.3	33.0	nr	29	Korsaeth et al., 2003
Turfgrass/high irrigation	Lysimeter	859†††	Sandy	–	433‡‡‡	38	45	nr	46	Barton et al., 2006
Turfgrass/high irrigation	Lysimeter	859	Sandy	433§§§	–	47	47	nr	50	Barton et al., 2006
Turfgrass/low irrigation	Lysimeter	859	Sandy	–	433	20	7	nr	74	Barton et al., 2006
Turfgrass/low irrigation	Lysimeter	859	Sandy	433	–	33	7	nr	83	Barton et al., 2006
Fruit trees	Suction cup	2500	Clay	–	100	1.2¶¶¶	1.3	nr	48	Renck and Lehman, 2004
Cereals	Tile drain	842###	Silty-clay-loam	240	–	7	52	nr	12	Murphy et al., 2000
Cereals	Tile drain	842	Silty-clay-loam	–	216	1.8	17.8	nr	9	Murphy et al., 2000
Cereals	Tile drain††††	775‡‡‡‡	Silty-clay-loam	–	88	0.6§§§§	48	nr	1	Lawes et al., 1881
Grassland	Suction plate	587	Sandy-loam/silty clay	0	0	0.9¶¶¶¶	1.3	nr	41	Oelmann et al., 2007
Grassland	Basin	nr	Clay	0####	0	6.2	2.7	108	70	Frank et al., 2000

† Calculated as $[DON/(DON + NO_3)] \times 100$.
‡ Losses averaged across 3 yr.
§ Across tillage and earthworm treatments.
¶ Average of 1, 8, and 9-yr clover sward.
# nr is not reported.
†† Calculated from Table 2 in Saarijarvi et al. (2004).
‡‡ Across CO_2 treatments.
§§ Average across 2 yr.
¶¶ Applied as urine, and mimicking a urine patch.
Applied as dung and mimicking a dung patch.
††† High irrigation is equal to 140% of daily replacement of pan evaporation; low irrigation is equal to 70% of daily replacement of pan evaporation.
‡‡‡ Average for water soluble and control-release fertilizer.
§§§ Average for pelletised poultry manure and biosolids.
¶¶¶ At 2 m depth.
Obtained from the Rothamsted archives.
†††† Based on data collected from a drain-gauge placed at 150 cm depth.
‡‡‡‡ Average rainfall between 1873 and 1879.
§§§§ Average based on 4 yr (1877–1881).
¶¶¶¶ Average of seven different levels of species richness and five different functional groups of grasses, small herbs, tall herbs, and legumes.
Grassland was in fallow for 15 yr at the initiation of the experiment.

excluded from the database (Wachendorf et al., 2005), the mean value for DON losses was 8.4 kg N $ha^{-1}yr^{-1}$, with a median value of 3.9 kg N $ha^{-1}yr^{-1}$. With the exception of a species-diverse grassland with low soluble N (DON and inorganic N) losses (Dijkstra et al., 2007) and a heavily fertilized turfgrass system with high soluble N losses (Barton et al., 2006), all other systems showed higher NO_3^- than DON losses. Across all agricultural systems, the mean loss of NO_3^- was calculated at 60.0 kg N ha^{-1} yr^{-1}, with a median value of 17.8 kg N $ha^{-1}yr^{-1}$. Higher leaching losses of NO_3^- compared to DON are expected, as NO_3^- is highly soluble and not bound by clay minerals like NH_4^+ (Feigenbaum et al., 1994). Moreover, net-N mineralization of organic matter leads to the production of NO_3^- as the conversion of NH_4^+ to NO_3^- occurs rapidly (Malhi and McGill, 1982).

On average, DON losses accounted for 26% of the total soluble N loss with a median value of 19% (Table 1). In other words, the amount of N lost as leached DON from a diverse set of agricultural systems, was estimated to be approximately one-third of the leaching losses observed for NO_3^-. Jiao et al. (2004) used intact 20 cm long intact soil cores collected from a no-till, conventional tilled field under maize (*Zea mays* L.) or soybean [*Glycine max* (L.) Merr.] which had received organic and inorganic fertilizer. They were placed in a laboratory setting and leached with synthetic rainwater. Dissolved organic N leaching ranged between 23 and 56% of the total N load with an average DON and NO_3^- loss estimated at 27 and 30 kg N ha^{-1}, respectively. As stated by Jiao et al. (2004) it would be difficult to predict from this experimental setting the nutrient load into the groundwater from agricultural practices. However, in relative terms, in this controlled leaching study DON losses were significant and comparable to NO_3^- losses.

Many biogeochemical models used to predict N leaching losses in agriculture have been focused solely on NO_3^- leaching (Andrews et al., 1997; Gerke et al., 1999; Garnier et al., 2001; Farahbakhshazad et al., 2008). From this survey, it is clear that DON leaching losses from agricultural fields can be a significant component of total N losses. Therefore, it should not be ignored when total N budgets are made. When biogeochemical models are updated to predict N losses, we suggest that a DON loss component should be included (Korsaeth et al., 2003).

In addition to DON, 4 of the 16 studies reported DOC losses which ranged between 30 and 174 kg C $ha^{-1}yr^{-1}$ (Table 1). The ratios of dissolved C to N ranged from 3 to 24. The DON and DOC pools are closely linked since similar organic compounds make up the DON and DOC pools and they are derived from the same organic matter pool. Part of the DOC can serve as a readily available substrate for soil microorganisms, leading to an increase in the mineralization of DON and subsequently nitrification (Brye et al., 2001). The DOC concentration in the soil solution can also impact the rate of denitrification, and therefore the concentration of DON (Burton and Beauchamp, 1985).

Concentration of Dissolved Organic Nitrogen in Leachates

Approximately half of the studies reported concentrations of DON in the leachate, collected at depths between 0.45 and 1.5 m. When DON concentrations in leachates were collected from more than one depth, only the concentrations of the leachates collected from the lowest depth are reported here. If in the study a treatment comparison was made, the lowest depth for which both DON and NO_3^- was provided for the treatments was chosen. Concentrations of DON ranged between 0.2 and 3.5 mg N L^{-1}. The lowest average concentration was found in the leachate collected from a cabbage field whereas the highest concentration occurred in a maize/soybean field under ridge tillage (Table 2). The allowable concentration in the EU of Kjeldahl N (organic N plus NH_4^+) in drinking water was reported at 1.0 mg per L^{-1} (European Community, 1980). In the United States, there is not a country-wide allowable standard for Kjeldahl N content in water. Instead, "criteria recommendations" are provided by eco-regions. For example, the criteria recommendation for Kjeldahl N content in rivers and streams for the western part of the United States, Ecoregion II, is 0.12 mg L^{-1} (http://www.epa.gov/waterscience/criteria/nutrient/ecoregions/rivers/rivers_2.pdf). Of the leachates collected from the various cropping systems, 90% showed a DON concentration (both mean and median values) that surpassed the allowable concentration for drinking water in the EU. Mean and median NO_3^- concentrations ranged between 0 and 18.6 mg N L^{-1}. Values above 11.3 and 10.0 mg of NO_3–N L^{-1} in the EU and United States, respectively, will exceed the allowable concentration for drinking water quality. Based on these allowable concentrations (mean and median), 17% of the leachates collected from the different cropping systems showed NO_3^- concentrations higher than the allowable concentration for drinking water quality. The high percentage of leachates samples which showed DON concentrations above the allowable concentration for drinking water was unexpected.

Some caution is required in interpreting these findings. First of all, the size of the data set is limited. Therefore, it is not clear how fully representative the data set is for DON and NO_3^- levels in leachates. Thus additional studies on DON concentrations in leachates in agricultural systems are required to confirm our findings. Second and with the exception of the Lawes et al. (1881) and Siemens and Kaupenjohann (2002) studies, the concentrations of DON were determined in leachate samples collected <1 m below the soil surface. It is likely that the DON concentrations in the solution would have decreased before the percolated water flowed into the drinking water basin or aquifer. This would occur via dilution from other sources of water or a reduction in DOC, and subsequently in DON, via microbial activity. For example, high denitrification potentials have been measured in the subsoil (Burton and Beauchamp, 1994; Van Groenigen et al., 2005), and denitrification would reduce DON and DOC concentrations in the soil solution.

Factors Controlling Leaching of Dissolved Organic Nitrogen

Leaching of DON occurs when water drains through the soil profile, with such events accentuated by the magnitude of the water flow and its duration as well as the duration of the antecedent predrainage period since the last time the soil was flushed (Cooper et al., 2007). Therefore, precipitation or irrigation events and their frequencies are likely the main drivers leading to DON losses in

Table 2. Concentrations of dissolved organic nitrogen (DON) and nitrate in leachates collected from agricultural systems. See Table 1 for further experimental details.

Cropping system	Depth	DON†	Nitrate‡	Reference
	m	mg N L^{-1}		
Ryegrass/maize	0.90	2.4	12.6	Siemens et al., 2003
Ryegrass/maize	0.90	2.6	18.6	Siemens et al., 2003
Fallow	0.90	1.8	9.0	Siemens et al., 2003
Fallow/sheep	0.90	1.5	5.8	Siemens et al., 2003
Maize/soybean/chisel till§	0.45	1.6	2.9¶	Shuster et al., 2003
Maize/soybean/chisel till#	0.45	3.2	6.8¶	Shuster et al., 2003
Maize/soybean/ridge till§	0.45	2.6	4.6¶	Shuster et al., 2003
Maize/soybean/ridge till#	0.45	3.5	6.0¶	Shuster et al., 2003
Cabbage	0.80	0.2††	2.5	Moller et al., 2005
Grass-clover/sandy	0.90	1.4	0–10	Vinther et al., 2006
Grass-clover/coarse sand	0.7	1.2–3.1	–	Vinther et al., 2006
Grassland-mono, low N‡‡	0.60	0.7	7.2	Dijkstra et al., 2007
Grassland-mono, high N‡‡	0.60	1.2	13.5	Dijkstra et al., 2007
Grassland-diverse, low N‡‡	0.60	1.6	1.1	Dijkstra et al., 2007
Grassland-diverse, high N‡‡	0.60	1.8	0.8	Dijkstra et al., 2007
Turfgrass, low irrigation/manure-N§§	0.98	–	–	Barton et al., 2006
Turfgrass, low irrigation/inorganic N§§	0.98	–	–	Barton et al., 2006
Turfgrass, high irrigation/manure-N§§	0.98	–	–	Barton et al., 2006
Turfgrass, high irrigation/inorganic-N§§	0.98	–	–	Barton et al., 2006
Pasture¶¶	NA##	1.6†††	–	Watson et al., 2000
Pasture‡‡‡	NA##	1.1†††	–	Watson et al., 2000

† Values above 1.0 mg L^{-1} of Kjeldahl N (organic N plus NH_4^+) exceeds the allowable concentration in the EU for drinking water quality.
‡ Values above 11.3 NO_3^-–N L^{-1} in the EU and 10 mg of NO_3^-–N L^{-1} in the United States exceeds the allowable concentration for drinking water quality.
§ Ambient earthworm population, average across years and management phases.
¶ Value include NO_2^- and NH_4^+.
# 100 earthworms per m^2 added; averaged across years and management phases.
†† Values calculated from Tables 4 and 5 in Dijkstra et al. (2007).
‡‡ Across CO_2 treatments.
§§ Averaged across two manure or two inorganic N treatments. Median values based on at least 138 values.
¶¶ Average across five rates of N fertilizer applications.
Depth not applicable as drainage water samples were collected at a weir.
††† Total Kjeldahl N (organic N plus NH_4^+).
‡‡‡ Average annual flow-weighted mean concentrations of six river catchments.

agricultural soils (Fig. 1). Even if large quantities of DON had accumulated in the top horizon of the soil profile, without a significant precipitation or irrigation event, leaching would not occur and no or limited losses of DON via leaching would take place. Rewetting the soil after a dry period, when the water content has been too low for mineralization and nitrification to take place, has also been shown to cause an increase in DON and DOC concentrations (Stark and Firestone, 1995; Lundquist et al., 1999).

The main sources of DON in agricultural soils are crop residues and soil organic matter, with DON being formed as part of the decomposition process. How much DON will be formed is dependent on a large number of agricultural management practices. Little is known on the effect of crop species or rotations on the concentrations of soluble soil N (Chantigny, 2003). However, Oelmann et al. (2007) found that the number of different species had little effect on DON losses but the presence of legumes led to an increase in DON losses.

As an increase in the quantity of crop residues or a change from summer fallow practices to a continuous cropping system increased soluble organic C (Campbell et al., 1999a,1999b; Graham et al., 2002), it is plausible that there would be a concurrent increase in soluble organic N which can lead to an increase in DON in the leachates following a precipitation event. Total DON leaching losses increased from a maximum of 4.7 kg N $ha^{-1}yr^{-1}$ under fallow to 9.2 kg N $ha^{-1}yr^{-1}$ when cropped with ryegrass (*Lolium* spp.) and maize (Siemens and Kaupenjohann, 2002; Table 1). An opposite result was observed by Vinther et al. (2006) who found that under fallow systems, losses of DON were higher than when cover crops were grown. However, as DOC losses remained the highest when a crop was present, the apparent contradictory result may have been caused by an earlier high percolation event. Application of manure also led to an increase in DON leaching (Murphy et al., 2000: Table 1). As pelletized poultry manure but not pelletized biosolids led to higher DON losses, not all organic amendments will lead automatically to higher DON losses (Barton et al., 2006).

Application of inorganic fertilizer N (40 kg ha^{-1} yr^{-1}) to a pasture composed of either single grass species or 16 different grass species led to an increase in DON losses but losses were higher with the higher number of species (Dijkstra et al., 2007, Table 1). It is possible that with the increase in the number of species, it may have proportionally increased the input of plant material and hence the source of DON. Increased DON losses when multiple species are present may also have been caused by differences among species and the effect of plant composition on DON leaching. Different grass species with different root phenology may cause different rates of leaching.

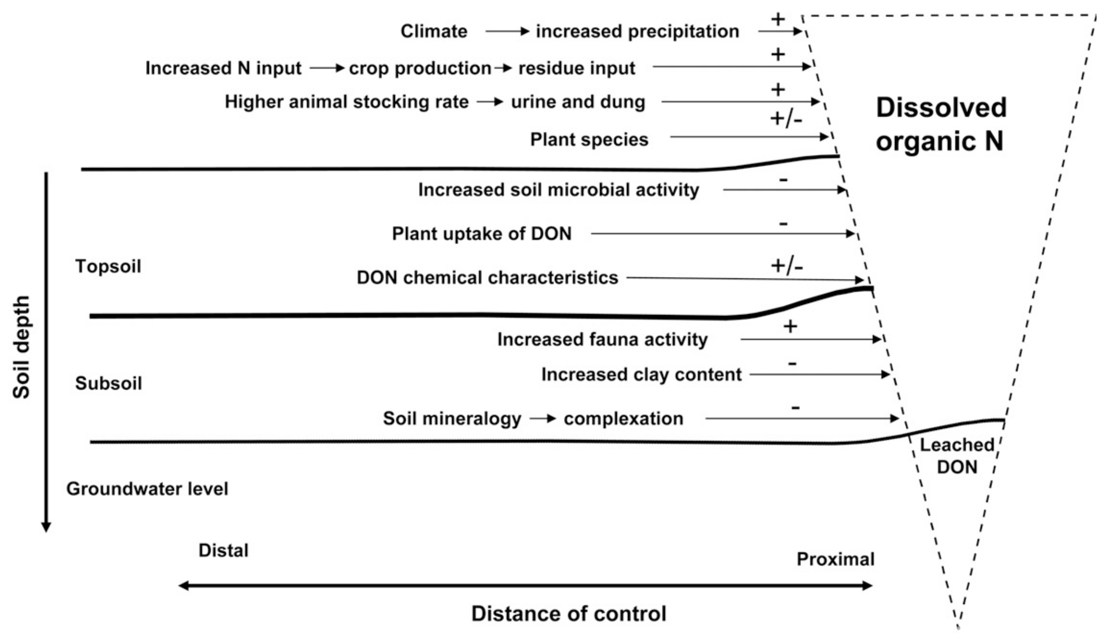

Fig. 1. Distal and proximal biophysical factors controlling the intensity of dissolved organic nitrogen (DON) leaching in agricultural fields.

Of interest is the finding that the addition of earthworms led to an increase in DON losses (Shuster et al., 2003; Table 1). Following the addition of 100 mostly anecic earthworms per m^{-2}, both DON and NO_3^- leaching losses increased significantly compared to ambient concentrations of earthworms. As anecic earthworms form semi-permanent vertical deep burrows, the presence of preferential leaching pathways may have been the cause of the increase in DON leaching.

The N content in urine can vary widely but is normally in the range of 8 to 15 g L^{-1} (Haynes and Williams, 1993). The amount of N under a cow (*Bos taurus*) urine patch can be equivalent to an application rate of 700 to 1200 kg N ha^{-1} (Jarvis et al., 1995); much higher than the demand of N for any agricultural crop. Urine also increases the pH of the soil following the hydrolysis of urea, the main form of N in urine (Haynes and Williams, 1993; Shand et al., 2002). The increase in soil solution pH often ranges over several units and can reach values of up to 9. This might considerably increase the DON and DOC content, as well as other compounds in the soil solution (Shand and Coutts, 2006). Urine patches become truly hotspots with regards to nutrient cycling and losses. Total amounts of N leached from urine patches were found to vary between 18 and 58% of the N applied (Clough et al., 1998). These highly localized concentrations of soluble N in combination with the effect of urine on solubilizing soil organic matter, lead to the highest DON leaching losses recorded with DON losses equivalent to up to 127 kg ha^{-1} yr^{-1} (Wachendorf et al., 2005; Table 1). The stocking rate, that is, the number of animals per hectare, will have a strong effect on the amount of urine deposited and subsequently DON losses.

Changes occur in the concentration and composition of DON as it moves through the soil profile. Dissolved organic N is used as a substrate by soil microbes. As DON is composed of different labile and more stable fractions, some fractions will be preferentially metabolized and the DON composition will change as it moves through the soil profile (Lajtha et al., 2005). Microbial consumption of labile, hydrophilic dissolved organic matter, that is, DON and DOC, will occur more rapidly than hydrophobic dissolved organic matter. In addition, hydrophobic dissolved organic matter with higher C-to-N ratios is more likely to show preferential sorption than hydrophilic dissolved organic matter, altering the DON composition at lower depths. These two processes lead to a generally observed decrease in the concentration of DON by depth in a wide array of forested and cultivated ecosystems (Lajtha et al., 2005; Moller et al., 2005; Renck and Lehmann, 2004; Vinther et al., 2006).

A third process that can lead to a decrease in the DON concentration as it moves through the soil profile is through the uptake of DON by plants. Using a double labeled ^{13}C-^{15}N amino acid commonly present in the soil, that is, glycine, the labels were detected in the shoot material of species present in a seminatural (*Festuca-Agrostis-Galium*) and improved grassland (*Lolium-Cynosurus*) (Streeter et al., 2000). The uptake of the ^{13}C-^{15}N amino acid occurred within 3 d following its application. Under conditions of limited N availability, these species did not show a preference for glycine-N or ammonium N as their source of N. When plants commonly used in grassland in northern Europe (timothy [*Phleum pratense* L.], alsike clover [*Trifolium hybridum* L.], red clover [*T. pretense* L.], and tall buttercup [*Ranunculus acris* L.]) were fertilized with combinations of ^{13}C-^{15}N labeled glycine, $^{15}NH_4^+$ and $^{15}NO_3^-$, all plants took up glycine in its intact form (Nasholm et al., 2000). As soluble organic N concentrations in agricultural soil are high and can be as high as inorganic N levels (Nemeth et al., 1988; Murphy et al., 2000; Bhogal et al., 2000; Jones and Willett, 2006), it has been used as justification for organic N being a source of crops in agricultural systems (Nasholm et al., 2000). The uptake of organic N was found to be widespread among many species from diverse ecosystems and to consist of an important source of N, in particular in ecosystems where microbial biomass is prone to large seasonal fluctuations and contributes to the release of labile organic N (Lipson and Nasholm, 2001). Jones et al. (2005) concluded that if DON is taken up by plants, it would still be premature to conclude that it is an important pathway of N uptake.

Most of the DON leaching studies were performed on light textured soils (Table 1). As light textured, sandy soils are known to be susceptible to high N leaching losses, it was likely to be the reason these soils were selected for the various studies. From this data set it would not be possible to conclude that soil texture and DON leaching losses were highly correlated as the texture of the various soils was limited to sandy or sandy-loam soils. Nevertheless, from NO_3^- leaching studies it is evident that sandy soils are more prone to leaching losses than clay soil (Clough et al., 1998; Arheimer and Liden, 2000). Therefore, it is likely that light textured soils are also more prone to DON leaching losses.

Conclusions

Although only a limited data set is published on DON losses from agricultural soils, every study which determined DON losses showed that N was lost as DON. In general, DON losses increased with increasing rates of inorganic and organic N applications. In particular following urine application to pastures or when high rates of organic and inorganic N were applied to turfgrass, DON losses became significant. It is evident that agricultural management practices cause DON losses to occur. With an average leaching loss of DON equal to a third of the NO_3^- losses, DON losses should be taken into consideration when total N budgets are constructed. As almost all of the leachates collected from agricultural fields exceeded the "criteria recommendations" of DON in drinking water in the United States, DON leaching losses can also pose a potential health hazard.

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