Nitrogen transformations in different types of soil treatment areas receiving domestic wastewater

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A B S T R A C T

Removal of N within the soil treatment area (STA) of onsite wastewater treatment systems is attributed to heterotrophic denitrification, with N lost to the atmosphere as N_2. However, the evidence supporting heterotrophic denitrification as the sole process for N removal is scant. We used $^{15}NH_4^+$ to follow N transformations in intact soil mesocosms representing a conventional STA receiving anoxic, C-rich wastewater, and two shallow-placed STAs receiving partially oxygenated, low-C wastewater. Nitrogen losses in the gas phase took place almost exclusively as $^{15}N_2$ in all STA types. We observed 10^2–10^3 times higher flux of N_2 than N_2O in all STAs, as well as net production of $^{15}N_2$ and $^{15}N_2O$ near the infiltrative surface and at greater depths in the soil profile. *In situ* net production of $^{15}NH_4^+$ suggested internal recycling of inorganic N in all STAs. The constraints imposed by differences in availability of electron donors and acceptors and soil physicochemical parameters in different STAs, point to autotrophic N removal processes (*e.g.*, anaerobic ammonia oxidation, autotrophic denitrification) as playing an important role in N removal. Our results suggest that N removal occurs at all depths of the STA, with losses due to both autotrophic and heterotrophic processes. Optimization of autotrophic N removal processes in the STA warrants further research efforts and may provide improved N removal.

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1. Introduction

The soil treatment area (STA; also referred to as drainfield or leachfield) of onsite wastewater treatment systems (OWTS; also referred to as septic systems) is optimized for wastewater infiltration and removal of pathogens, but not N removal. Increases in the size and density of populations served by OWTS have caused higher N concentrations in receiving ground and surface water (Valiela et al., 1992). Elevated N inputs to aquatic environments cause eutrophication of coastal and fresh water ecosystems (Howarth and Marino, 2006), and negatively impacts human health (Oakley et al., 2010).

Nitrogen removal does take place in the STA, with rates ranging from 0 to 51% (Bunnell et al., 1999; Siegrist et al., 2014; USEPA, 2002). However, the processes involved are poorly understood. Heterotrophic denitrification, which produces N_2 and N_2O, is generally considered to be responsible for N removal (Bradshaw and Radcliffe, 2013; Bunnell et al., 1999; Crites and Tchobanoglous,

1998), and is thought to occur near the infiltrative surface. Conditions that support N removal by heterotrophic denitrification are likely to prevail in conventional STAs, which receive periodic inputs of anoxic effluent with high organic C and NH_4^+ levels, followed by oxic periods during which NO_3^- can be produced. Heterotrophic denitrification is less likely in a shallow-placed, or advanced, STAs because wastewater inputs have higher dissolved oxygen (DO) levels and much lower organic C levels due to rapid biodegradation in pre-treatment steps.

Processes other than heterotrophic denitrification can result in N removal in gaseous forms, including nitrification (N_2O), anaerobic ammonia oxidation (N_2), and autotrophic denitrification (N_2 and N_2O) (Table 1). Attributing all N removal in the STA to heterotrophic denitrification may be an over simplification of the processes involved. Autotrophic denitrifiers can reduce NO_3^- to N_2 using reduced S compounds (Kanter et al., 1998; Robertson et al., 1991Robertson and Kuenen, 1991) or CH_4 (Islas-Lima et al., 2004) as an electron donor, both of which are present in wastewater. Production of N_2 via anaerobic oxidation of NH_4^+ with either NO_2^- (anammox) (Robertson et al., 2012; Strous et al., 1997) or Fe^{3+} (feammox) (Yang et al., 2012) is also possible, with NO_2^- produced *in situ* and Fe^{3+} produced in soil. Loss of N as N_2O can also occur

Table 1

Minimal processes that can contribute to production of N_2 and N_2O in the soil treatment area of onsite wastewater treatment systems (OWTS).

Process	C source	Electron donor	Electron acceptor	Relative production of N_2 and N_2O	References
Heterotrophic denitrification	Organic	Organic	NO_3^-	$N_2 > N_2O$	Bradshaw and Radcliffe, 2013; Bunnell et al., 1999; Crites and Tchobanoglous, 1998
CH_4-coupled denitrification	CO_2	CH_4	NO_3^-	N/A[*]	Islas-Lima et al., 2004
S-coupled denitrification	CO_2	H_2S, S^0	NO_3^-	$N_2 \gg N_2O$	Kanter et al., 1998; Robertson and Kuenen, 1991
Anaerobic NH_4^+ oxidation	CO_2	NH_4^+	NO_2^-	N_2 only	Robertson et al., 2012; Strous et al., 1997
Fe-coupled NH_4^+ oxidation	CO_2	NH_4^+	Fe^{3+}	N_2 only	Yang et al., 2012

[*] Not available.

from autotrophic nitrification (Wrage et al., 2001). These processes all have different requirements in terms of C, electron donors, electron acceptors and redox conditions that can be met in different niches within the STA.

An improved understanding of N transformations in the STA, particularly those that result in gaseous N losses, can help optimize N removal. In a previous study, we observed 12.0% N removal from a conventional STA receiving septic tank effluent, and 4.8–5.4% N removal from two advanced STAs receiving wastewater that had been treated in a single-pass sand filter (Cooper et al., 2015). In the present study, we examined the transformations of N in these STAs using wastewater amended with $^{15}NH_4^+$ to better understand the type and location of N removal processes within the STA. We hypothesized that autotrophic processes would contribute to gaseous N losses in addition to heterotrophic processes, and that removal would take place at greater depths within the soil profile as well as near the infiltrative surface. We measured aqueous ($^{15}NH_4^+$ and $^{15}NO_3^-$) and gaseous ($^{15}N_2$ and $^{15}N_2O$) species over time and within the soil profile of the STA. In addition, we quantified variables that may affect N transformations, including BOD_5, pH, SO_4^{2-}, CH_4, CO_2, O_2, and water-filled pore space (WFPS).

2. Materials and methods

2.1. Experimental setup

We used intact soil mesocosms (152-cm tall × 15-cm-dia.) to represent three STA types: (i) conventional pipe and stone (P&S), (ii) shallow narrow drainfield (SND) and (iii) Geomat® (GEO), a SND variation (Supplemental Fig. S1) in accordance with the design and dosing regulations set by the Rhode Island Department of Environmental Management (RIDEM, 2013). The P&S was dosed with 200 mL of septic tank effluent (STE) every 12 h over 1.5 h, corre-

Table 2

Characteristics of septic tank effluent (STE) for the pipe and stone (P&S) soil treatment area, and sand filter effluent (SFE) for shallow narrow (SND) and Geomat® (GEO) soil treatment areas used in this study. Units are mg L^{-1} except for electrical conductivity (μS/cm) and pH. (n = 1).

Property	P&S		SND		GEO	
	Input	Output	Input	Output	Input	Output
pH	6.9	3.7	5.6	3.4	5.4	3.5
Dissolved oxygen	0.1	0.0	3.1	2.0	3.4	0.6
5-day biochemical oxygen demand	190	0.0	4.0	0.0	12	0.0
Total suspended solids	46	0.0	2.0	0.0	2.0	0.0
Electrical conductivity	880	750	470	560	470	550
Total N	74	70	54	47	61	41
NH_4^+-N	53	0.10	15	2.5	14	3.1
NO_3^--N	0.05	57	10	32	27	38
SO_4^{2-}-S	7.2	8.4	29	9.4	21	8.8

sponding to 400 mL d^{-1} (22.6 L m^{-2} d^{-1}). The SND and GEO received wastewater that had passed through a single-pass sand filter (SFE). They were dosed with 22.5 mL SFE every 30 min over 15 min, corresponding to 2000 mL d^{-1} (113 L m^{-2} d^{-1}). Septic tank effluent and SFE were collected weekly from the same treatment train at a residence in Charlestown, RI, USA. Characteristics of wastewater inputs can be found in Table 2, and were within the range of those reported previously (Cooper et al., 2015). Further details of the experimental design, sampling, and analytical methods can be found in Cooper et al. (2015).

2.2. ^{15}N tracer experiment

The experiment was conducted between 7 January and 8 February 2014, after the soil mesocosms had been in continuous operation for 52 weeks. ^{15}N-labeled ammonium chloride ($^{15}NH_4Cl$, ~98+ At.% (that is, ~98 N atoms of 100 are labeled ^{15}N), Isotec Chemical Co., Miamisburg, OH) was added to either STE (for P&S) or SFE (for SND and GEO). We added 420 μg, 482 μg, and 391 μg of ^{15}N to P&S, SND and GEO, respectively, resulting in final concentrations of 2.1, 3.7, and 3.1 mg ^{15}N L^{-1}. Wastewater amended with ^{15}N was delivered to the P&S drainfield in a single, 200-mL dose pumped over 1.5 h. Delivery of ^{15}N-amended wastewater to the SND and GEO drainfields was achieved by dosing with 126 mL (three, 42-mL doses every 30 min for 15 min) over 1.5 h.

2.3. Analysis of ^{15}N in aqueous and gas phase

Water samples for $^{15}NH_4^+$ and $^{15}NO_3^-$ analyses were diffused using to the acidified disk diffusion method (Khan et al., 1998) at 20 °C using 10 mL samples. Diffusates and gases were analyzed at the University of California-Davis Stable Isotope Facility. Diffusates were analyzed with a continuous flow isotope ratio mass spectrometer (PDZEuropa, Northwich, UK) after combustion of samples at 1000 °C to convert samples to N_2 with an online elemental analyzer (PDZEuropa, ANCA-GLS) according to Mulvaney (1993). Concentrations of $^{15}N_2$, $^{15}N_2O$, N_2 and N_2O were determined by gas chromatography (PDZEuropa, TGII trace gas analyzer) followed by a dual inlet isotope ratio mass spectrometer (PDZEuropa 20-20 IRMS) to attain separation of N_2 and N_2O (Mosier and Schimel, 1993).

2.4. ^{15}N calculations

Natural abundance values, determined on samples taken prior to the start of the experiment, were subtracted from all enriched