Mechanisms of Ammonium Transformation and Loss in Intermittently Aerated Leachfield Soil

José A. Amador, Laboratory of Soil Ecology and Microbiology, University of Rhode Island, Kingston, RI (jamador@uri.edu)
John T. Richard, Rhode Island NRCS, Warwick, RI
David A. Potts, Geomatrix LLC, Old Saybrook, CT

Background

- Conventional onsite wastewater treatment systems (OWTS) can degrade ground, surface and coastal water quality from excess N inputs in rural and suburban areas.
- Nitrogen removal from wastewater is not the primary function of conventional OWTS, and removal rates in the soil vary from 0 to 30% (U.S. EPA, 2002).
- In the absence of direct mechanistic information, N removal in conventional drainfields is attributed to denitrification (e.g. Crites and Tchobanoglous, 1998), which produces N_2 and N_2O – gases that are readily lost to the atmosphere.
- A better understanding of the specific mechanisms involved may help to optimize system conditions to improve N removal.
- We have shown that intermittent aeration of a conventional leachfield receiving domestic septic tank effluent (STE) achieves N removal rates of 25% to 75% (Potts et al., 2004; Amador et al., 2007; Amador et al., 2010).
- We have hypothesized that intermittent aeration enhances N removal by promoting nitrification (during the aeration phase), followed by denitrification (as STE infiltrates and percolates through the soil), with net N losses attributed to N_2 and N_2O produced by denitrification (Potts et al., 2004).
- In the present study we used $^{15}NH_4^+$-amended STE to test this hypothesis by tracking the transformations and fate of ^{15}N within intermittently aerated leachfield mesocosms.

Experimental

Mesocosms

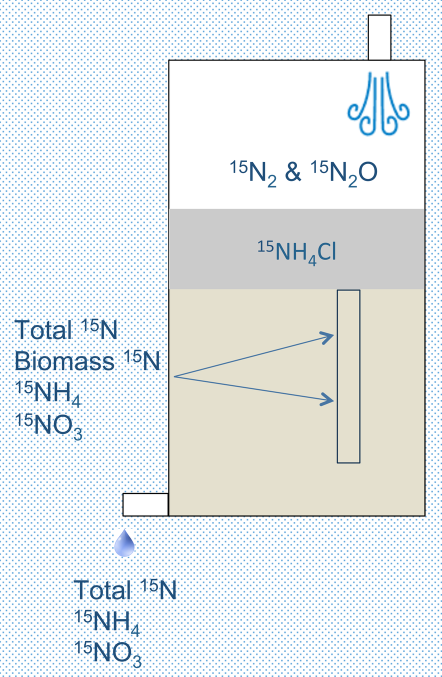
- Replicated (n = 3) stainless steel mesocosms (36.0-cm internal dia., 61.0-cm high) filled with 30 cm sandy loam and 7.5 cm silica sand.
- Dosed with STE every 6 h at 12 cm d^{-1}
- Air was pumped for 30 min into the headspace of the mesocosms using a blower to maintain ambient O_2 levels (0.20 to 0.21 mol mol^{-1}), followed by a 60-min period with the blower off.

^{15}N Addition

- $^{15}NH_4Cl$ (~ 98+ At.%) was mixed with 22.9 L of STE to a final concentration of 1 mg ^{15}N L^{-1} (~ 3.8 mg $^{15}NH_4Cl$ L^{-1}) and each mesocosms dosed manually once with 2.8 L of ^{15}N-amended effluent.

Sampling and Analyses

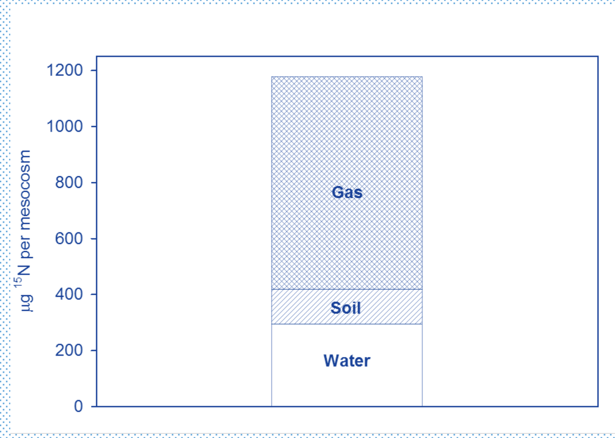
- Gas samples taken every 6 h
- Water collected for 24 h
- Soil cores (30-cm deep) taken 24 h after dosing
- Samples analyzed for labeled and unlabeled forms of N.



Acknowledgements

This study was funded by a grant from the National Oceanic and Atmospheric Administration/University of New Hampshire Cooperative Institute for Coastal and Estuarine Environmental Technology (CICEET) and by the Rhode Island Agricultural Experiment Station. We thank Barbara Nowicki, Erika Nicosia and Janet Atoyan for technical assistance. This is contribution no. 5408 of the Rhode Island Agricultural Experiment Station.

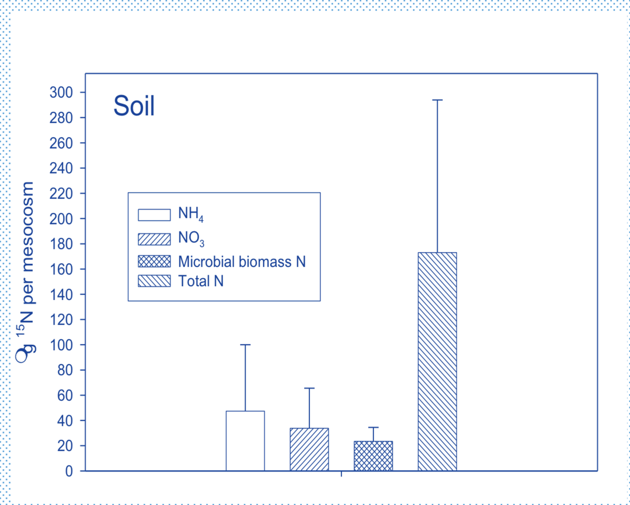
^{15}N Distribution – General



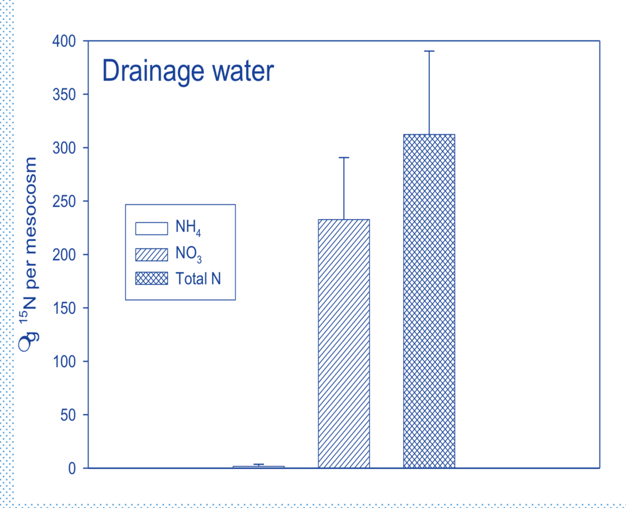
- Of the ~2,700 μg ^{15}N added to each mesocosm, we were able to account for ~1,200 μg, or 44.4%.
- Assuming that all of the unaccounted ^{15}N was in the gas phase, we estimate that, 24 h after addition of $^{15}NH_4^+$ to the mesocosms, 5.7% of the added ^{15}N was found in the soil, 10.0% in the drainage water, and 84.3% in the gas phase.

^{15}N Distribution – Soil

- The mass of ^{15}N associated with TN and inorganic N ($^{15}NH_4^+$ + $^{15}NO_3^-$) in soil was 120 and 85 μg ^{15}N $mesocosm^{-1}$, respectively.
- The mass of ^{15}N associated with extractable NH_4^+ and NO_3^- was 50 and 35 μg ^{15}N $mesocosm^{-1}$, respectively.
- ^{15}N in soil microbial biomass was a 9 μg ^{15}N $mesocosm^{-1}$, a minor fate for $^{15}NH_4$, accounting for 0.3% of added ^{15}N and 7.5% of the ^{15}N pool in soil.

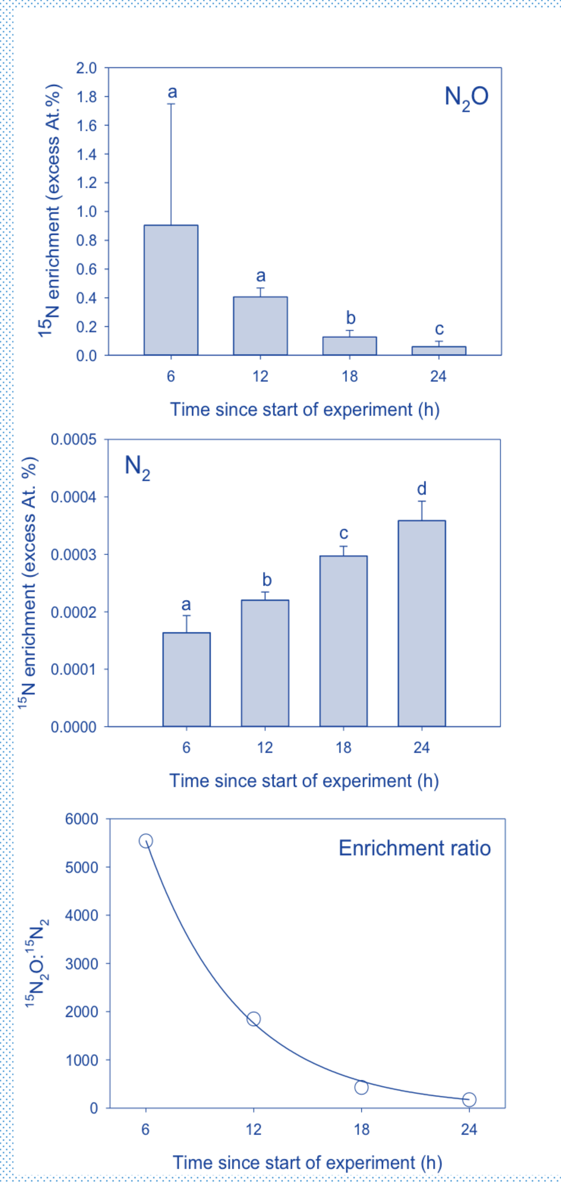


^{15}N Distribution – Drainage Water



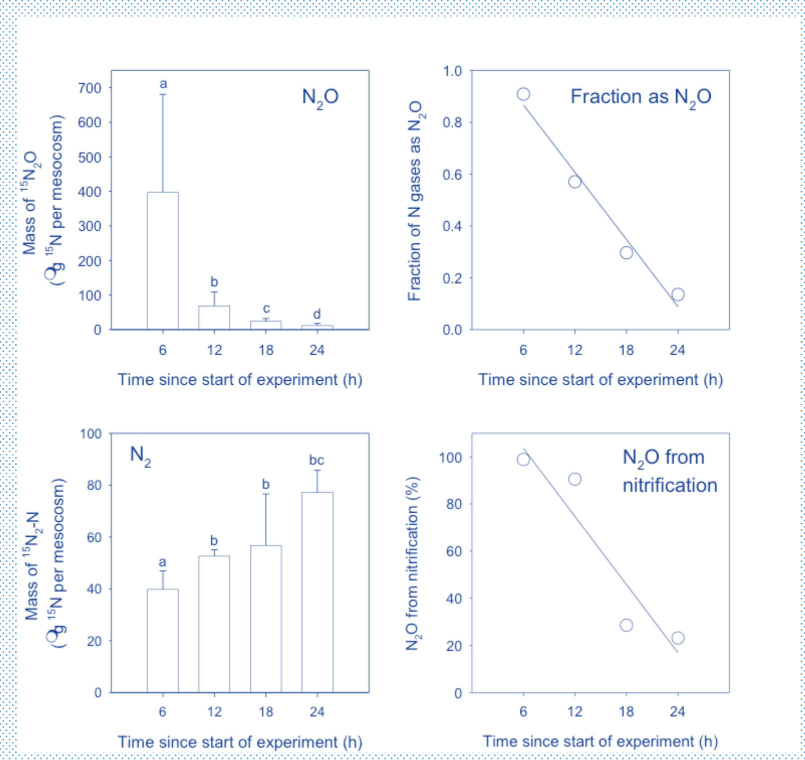
- The mass of ^{15}N in TN in drainage water was 314 μg ^{15}N $mesocosm^{-1}$.
- The inorganic N pool had a ^{15}N mass of 233 μg $mesocosm^{-1}$, nearly all of it as NO_3^- – nitrification is a major pathway for NH_4 transformation.
- Difference between total ^{15}N and the sum of $^{15}NH_4$ and $^{15}NO_3$ represents ^{15}N-labled organic N. This pool accounts for ~20% of the ^{15}N in drainage water, and include nitrogenous organic metabolites excreted by microorganisms, and ^{15}N-labeled live and dead microorganisms.

^{15}N Enrichment – Gases



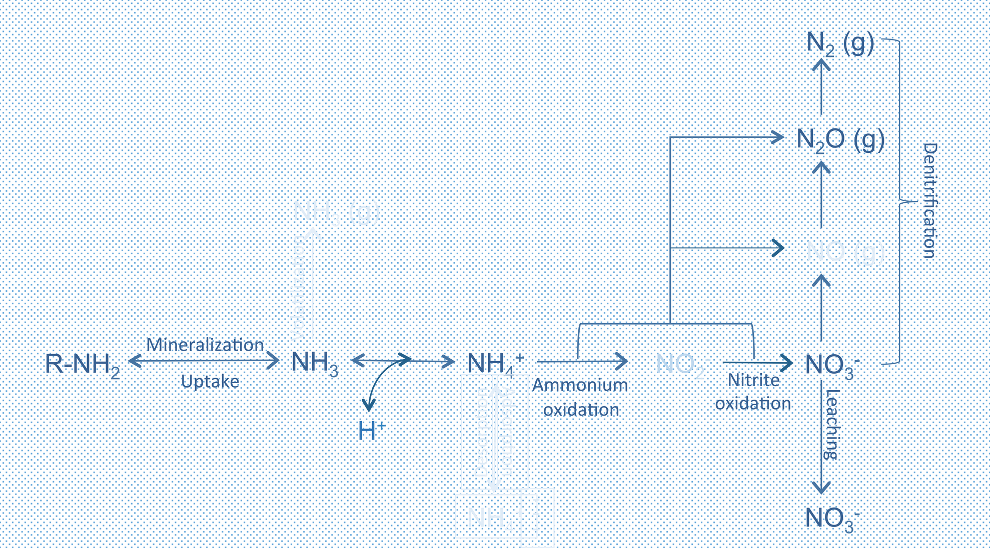
- Enrichment of N_2O with ^{15}N was highest after 6 h, decreasing exponentially thereafter.
- ^{15}N enrichment of the N_2 pool was considerably lower than for N_2O, and increased linearly throughout the experiment.
- Ratio of N_2O and N_2 enrichment with ^{15}N decreased exponentially with time.
- If production of $^{15}N_2$ and $^{15}N_2O$ occurs strictly via sequential nitrification-denitrification, and $^{15}N_2O$ is produced only by denitrification, we would expect (i) the concentration of both $^{15}N_2O$ and $^{15}N_2$ to increase with time as the concentration of $^{15}NO_3$ from oxidation of $^{15}NH_4$ increases, and (ii) the ratio of ^{15}N enrichment of N_2O and N_2 to remain constant throughout the experiment, since both gases would be formed from the same pool of $^{15}NO_3$. Our results do not support the hypothesis that denitrification is solely responsible for production of nitrogenous gases.
- Although sequential nitrification and denitrification are clearly taking place, as evidenced by production of $^{15}N_2$ from added $^{15}NH_4$, our results suggest that $^{15}N_2O$ production results both from denitrification and nitrification.

^{15}N Distribution – Gases



- The mass of $^{15}N_2O$-N in the headspace was highest at 6 h (~ 400 μg $^{15}N_2O$-N $mesocosm^{-1}$), declining exponentially to ~15μg $^{15}N_2O$-N $mesocosm^{-1}$ after 24 h.
- By contrast, the mass of $^{15}N_2$-N in the headspace increased linearly with time, from 40 μg $^{15}N_2$-N $mesocosm^{-1}$ after 6 h to a maximum of 80 μg $^{15}N_2$-N $mesocosm^{-1}$ after 24 h.
- The fraction of ^{15}N gases comprised by N_2O declined linearly with time, from a maximum of 92% at 6 h to a low of 15% at 24 h.
- We estimated the relative contribution of nitrification and denitrification to N_2O production using the approach described by Khalil et al. (2004), applying an N_2O-to-N_2 ratio of 0.12.
- The contribution of nitrification to the N_2O pool was highest at 6 h (98.8%), declining to 23.1% at 24 h.
- The alternating oxic and anoxic conditions in our mesocosms support nitrification and denitrification, both of which contribute to N_2O production, and to gaseous losses of N from these systems.

Proposed NH_4^+ Transformation Pathways



Summary

- Our results show that 5.7% of the ^{15}N applied as $^{15}NH_4Cl$ was found in soil, 10.0% in drainage water, and 84.3% was in the gas pool.
- Ammonium accounted for 41.7% of the soil ^{15}N pool, followed by NO_3^- (29.2%), organic N (21.7%), and microbial biomass N (7.5%).
- In drainage water, NO_3^- constituted ~80% of the ^{15}N pool, whereas NH_4^+ was absent from this pool.
- Nitrous oxide was the dominant form of ^{15}N in the gas phase 6 h after addition of $^{15}NH_4^+$-amended STE to the mesocosms, after which its mass declined exponentially; by contrast, the mass of $^{15}N_2$ was initially low but increased linearly with time to become the dominant form of ^{15}N after 24 h.
- Isotopic enrichment of $^{15}N_2O$ and $^{15}N_2$ indicates that nitrification contributed 98.8% and 23.1% of the $^{15}N_2O$ flux after 6 h and 24 h, respectively.
- Gaseous losses are the main mechanism for NH_4^+ removal from wastewater in intermittently aerated soil.
- **Nitrification – generally not considered a significant pathway for N loss in soil-based wastewater treatment – is an important source process for N_2O.**

References

Amador, J.A., D. A. Potts, G.W. Loomis, D. V. Kalen, E. L. Patenaude, and J. H. Görres. 2010. **Improvement of hydraulic and water quality renovation functions by intermittent aeration of soil treatment areas in onsite wastewater treatment systems.** *Water* **2**:886–903.

Amador, J.A., G.W. Loomis, D. Kalen, E.L. Patenaude, and J.H. Gorres. 2007. **Evaluation of Leachfield Aeration Technology for Improvement of Water Quality and Hydraulic Functions in Onsite Wastewater Treatment Systems**. Final Report – The NOAA/UNH Cooperative Institute for Coastal and Estuarine Environmental Technology (CICEET).

Crites, R., and G. Tchobanoglous. 1998. **Small and Decentralized Wastewater Management Systems.** McGraw-Hill, New York.

Khalil, K., B. Mary, and P. Renault. 2004. **Nitrous oxide production by nitrification and denitrification in soil aggregates as affected by O_2 concentration.** *Soil Biol. Biochem.* **36**:687–699.

Potts, D. A., J.H. Görres, E.L. Nicosia, and J.A. Amador. 2004. **Effects of aeration on water quality from septic system leachfields.** *J. Environ. Qual.* **33**:1828–1838.

USEPA. 2002. **Onsite Wastewater Treatment Systems Manual.** EPA/625/R-00/008. USEPA, Office of Water, Washington, DC.