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Partial nitrification enhances natural attenuation of nitrogen in a septic system plume

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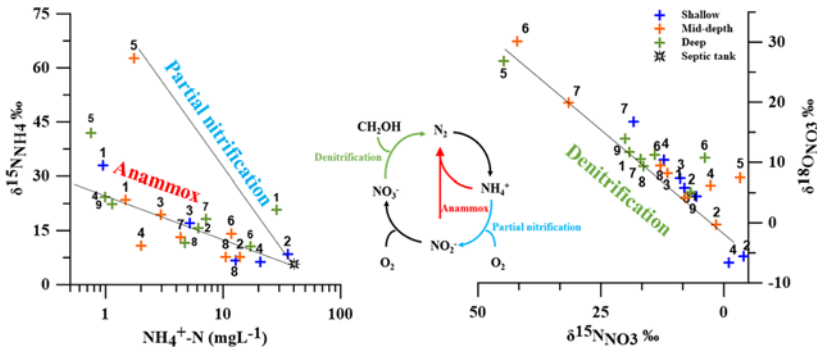
HIGHLIGHTS

- Up to 80% of N removal was observed at theKillarney plume with respect to the septic effluent due to natural attenuation.
- An isotopiccharacterization of both NH_4^+ and NO_3^- were used to provide insight into potential attenuation processes.
- Anammoxand denitrification contributed to the observed TIN attenuation.
- Field datademonstrate that N attenuation can be enhanced by controlling the water tabledepth in septic system plumes.

GRAPHICAL ABSTRACT



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ABSTRACT

Natural attenuation of nitrogen (N) was investigated in a well characterized septic system plume at a campground in Ontario, Canada. Total inorganic N (TIN) concentrations in deeper portions of the plume were about one third of the septic tank value of 40.7 mg L^{-1}. NH_4^+ and NO_3^- isotopic characterization were used to provide insight into potential attenuation processes. Concentrations of NH_4^+ and NO_3^- were highly variable in the plume, but approached the septic tank TIN value in some shallow zones and exhibited $\delta^{15}N$ values like the tank value of +6‰. However, isotopic enrichment (up to +24‰ for NH_4^+ and +45‰ for NO_3^-) and declining TIN concentrations in the deeper zones indicated that anaerobic ammonium oxidation contributed to the TIN attenuation. The degree of isotopic enrichment increased at lower NH_4^+ concentrations and was consistent with Rayleigh-type distillation with an enrichment factor (ε) of −5.1‰. Additionally, decreasing DOC values with depth and the concomitant enrichment of $\delta^{15}N_{NO3}$ and $\delta^{18}O_{NO3}$, suggested that denitrification was also active.
The N attenuation observed in the Killarney plume was partly due to incomplete nitrification that occurred because of the shallow water table, which varied from only 0.2–0.7 m below the tile bed infiltration pipes. Moreover, some of the monitoring locations with the shallowest water table distances from the infiltration pipes, had the highest degree of TIN attenuation (70–90%) in the plume. This behavior suggests that controlling water table distance from the infiltration pipes could be a useful mechanism for enhancing N attenuation in septic system plumes.

1. Introduction

Nitrogen (N) contamination of groundwater is a worldwide concern and on-site wastewater disposal in septic systems represents a

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potentially significant source of N loading to groundwater in some areas (Withers et al., 2014). Often, septic tank systems act as point-sources of contamination which can generate high local concentrations of both ammonium (NH_4^+), and when the wastewater is subsequently oxidized during infiltration, nitrate (NO_3^-) (Withers et al., 2014; Lawrence et al., 2000; Christensen et al., 2001; Heaton et al., 2005; Gooddy et al., 2014 and Groeschke et al., 2016). Although the primary concern of septic tank systems might be in relation to pathogenic pollutants (Macler and Merkle, 2000), N compounds can also represent a serious challenge, because if not attenuated, they can migrate large distances and impact receptors such supply wells and surface waters.

Therefore understanding the fate of N compounds disposed of in septic systems is vital for assessing risk to groundwater supplies and particularly because N compounds can be affected by a wide range of reactions during subsurface flow. For example, Richards et al. (2016) presented an assessment of multiple pollutants from thirty two septic systems in the UK, combining physical, chemical and microbial compositions of effluent from households, whereas a modeling approach was applied to simulates both ammonium and nitrate transport from septic systems to surface water bodies in the US (Zhu et al., 2016).

Despite the concern for NH_4^+ contamination of aquifers, only a few studies have focused on the potential for natural attenuation (Aravena and Robertson, 1998; Ptacek, 1998; Swartz et al., 2006; Robertson et al., 2013). Attenuation reactions can play an important role in degrading N compounds, thereby reducing the potential detrimental effects on the quality of groundwater, surface waters, related ecosystems and human health. In this framework, this study has high relevance for the better management and protection of water resources.

In the current study, chemical and isotopic tools were used to investigate of a well characterized septic system plume originating from a campground at Killarney Provincial Park, Ontario (ON, Canada), to address the following objectives: (i) examine the biogeochemical processes responsible for the N attenuation along the plume; (ii) demonstrate if partial nitrification enhances N removal by anammox, and (iii) investigate the role of the water table distance from the infiltration pipes as a contributing factor in N attenuation processes.

1.1. Nitrogen transformation in septic systems

Septic systems are a passive means for disposing of wastewater and contaminants undergo a variety of biochemical and geochemical reactions during subsurface disposal in septic systems (Spiteri et al., 2008). There are typically three major compartments in which wastewater is transformed: within the septic tank, during infiltration through the unsaturated zone and then in the septic system plume which develops in the groundwater zone. The septic tank usually represents an anaerobic environment, with low dissolved oxygen (DO), but with high organic matter content. Under such conditions and with adequate retention time, most N is released from organic molecules and is mineralized to inorganic NH_4^+ (Rivett et al., 2008). During transport through the unsaturated zone NH_4^+ may be transformed by nitrification (Eqs. (1), (2)). Here, microorganisms use O_2 to oxidize NH_4^+ to NO_3^- (Rivett et al., 2008). As a result, groundwater plumes with high NO_3^- concentrations may develop, or if nitrification is incomplete, plumes may contain both NH_4^+ and NO_3^-. If NH_4^+ and NO_3^- coexist in a plume, N attenuation can result from anaerobic oxidation of ammonium (anammox, Eq. (3)) or if an electron donor such as CH_2O is available, NO_3^- can be attenuated by denitrification (Eq. (4); Böhlke et al., 2006).

$$2NH_3{}^+ 3O_2 \rightarrow 2NO_2{}^- + 2H^+ + 2H_2O \quad (1)$$

$$2NO_2{}^- + O_2 \rightarrow 2NO_3{}^- \quad (2)$$

$$NH_4{}^+ + NO_2{}^- \rightarrow N_2 + 2H_2O \quad (3)$$

$$4NO_3{}^- + 5CH_2O \rightarrow 2\,N_2 + 4HCO_3{}^- + CO_2 + 2H_2O \quad (4)$$

The stable isotopes $\delta^{15}N_{NH4}$, $\delta^{15}N_{NO3}$ and $\delta^{18}O_{NO3}$ have been widely used to gain insight into N transformation processes in a variety of settings including in septic system plumes, starting at the shallow zone directly beneath the tile bed, and then along the groundwater flow system (Vitòria et al., 2008; Robertson et al., 2012). This case study contributes to a better understanding of geochemical reactions occurring within septic plumes, the factors controlling these reactions, and potential implications for future wastewater plumes management.

2. Materials and methods

2.1. Site description

Killarney Provincial Park (ON) hosts an overnight campground with 128 campsites that is open year-round. Wastewater loading is estimated to average 14 $m^3\ d^{-1}$ during the summer peak use period (May to October). All wastewater is discharged to a single septic system that has been in operation since 2004. The septic system includes a septic tank from which the wastewater is periodically pump-dosed to a filter bed, 2700 m^2 in area. The filter bed is constructed of imported, medium-coarse filter sand that overlies lacustrine clay at 2 m below ground (b.g.) depth (Fig. 1). In a previous study (Wen, 2013), the filter bed sand was assessed by electron microscopy with x-ray elemental analysis, which revealed that quartz and feldspar were the dominant mineral components and carbonate minerals (limestone and dolomite) were absent. Thus, the filter sand has low buffering capacity and water/rock interaction in the plume was expected to be relatively weak.

Groundwater forms a mound under the northern portion of the filter bed as a result of the wastewater loading and the low permeability of the underlying clay and then flows predominantly southward toward a small creek (average flow, several L min^{-1}) that skirts the perimeter of the filter bed (Fig. 1). As a result of this mounding, the water table was relatively shallow at the time of the study and varied from 0.2–0.7 m below the wastewater infiltration pipes (Fig. 1).

2.2. Field and analytical approach

The groundwater monitoring network at the site is comprised of 30 monitoring tubes, contained in 12 multilevel sampling bundles (nests); nine of the bundles located along the plume centerline were sampled in this study (KP1 ÷ 9, Fig.1). Sampling occurred during August 11–13, 2014 which represented the middle of the peak use period. All the multilevel wells are located within and up to 7 m beyond the edge of the tile bed. Groundwater was collected from the septic tank by a dedicated polyethylene sampling tube installed near the outlet of the tank, whereas groundwater samples were obtained from the multilevel bundle piezometers. These consisted of three sampling tubes each, set at varying depths (b.g.): shallow (s) from 1.2 to 1.4 m, mid-depth (m) from 1.5 to 1.7 m, and deep (d) from 1.8 to 2.0 m. During sample collection, electrical conductivity (EC), pH and dissolved oxygen (DO) values were determined in the field, prior to atmospheric exposure, using portable meters. Groundwater samples were collected using a peristaltic pump with silicone tubing. Samples were immediately filtered (0.45 μm Millipore filter) and were stored refrigerated at 4 °C, and in a dark environment until transported to the laboratory for proper conservation prior to analysis. Concentration and isotopic analyses were performed at Department of Earth and Environmental Sciences, University of Waterloo (ON) in the Environmental Geochemistry (EGL) and in the Environmental Isotope (EIL) Laboratories, respectively.

Nitrate and chloride concentrations were measured by ion chromatography using a Dionex ICS 2100 (Sunnyvale, CA) equipped with an IonPac AS 18 Analytical column (2 × 250 mm) and an AS 18 Guard Column (4 × 50 mm); the analytical error is 0.2 mg L^{-1}. Nitrite and ammonium were run on a SmartChem 200 Discrete Analyzer, Unity Scientific