



Changes to nitrate isotopic composition of wastewater treatment effluent and rivers after upgrades to tertiary treatment in the Narragansett Bay watershed, RI



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ABSTRACT

Due to nitrogen load reduction policies, wastewater treatment facilities (WWTFs) have upgraded to tertiary treatment — where denitrification reduces and removes nitrogen. Changes to the stable isotopic composition of nitrate inputs after upgrades or how it transfers to the estuary have not been assessed in Rhode Island. We investigate whether these upgrades impact the isotopic signature of nitrate inputs to Narragansett Bay. Samples from rivers and WWTFs discharging to Narragansett Bay characterize the anthropogenic source nitrate (NO_3^-) isotopic composition ($\delta^{15}\text{N}-\text{NO}_3^-$ and $\delta^{18}\text{O}-\text{NO}_3^-$) and temporal variability. At one WWTF, tertiary treatment increased effluent nitrate $\delta^{15}\text{N}-\text{NO}_3^-$ and $\delta^{18}\text{O}-\text{NO}_3^-$ values by ~16%. Riverine values increased by ~4%, likely due to the combination of decreases in N and upgrades. Combined river and WWTF flux-weighted isotopic compositions showed enriched values and an amplitude reduction in monthly variability. When seasonal isotopic means are significantly different from other sources, $\delta^{15}\text{N}-\text{NO}_3^-$ may be a useful tracer of inputs.

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1. Introduction

To reduce the negative effects of anthropogenic nitrogen (N) loads, namely eutrophication and hypoxia, some wastewater treatment facilities (WWTFs) include nitrogen removal in their treatment schemes (USEPA, 2004; RIDEM, 2005). During processing, wastewater is initially strained to remove solids and grease/oil from water to be treated. Next, secondary treatment converts dissolved organic matter to inorganic N and then oxidizes ammonium (NH_4^+) to nitrate (NO_3^-), a microbial process called nitrification. The newest step, tertiary treatment, reduces NO_3^- , through microbially-mediated denitrification, to the largely biologically unavailable N_2 gas. Finally, wastewater is disinfected and released. The entire process takes about 1 day (USEPA, 2004).

Addition of tertiary treatment has a dramatic effect on the final composition of wastewater. The Narragansett Bay Commission (NBC), the company that owns two of the largest WWTFs that discharge into Narragansett Bay, RI, upgraded their facilities to tertiary treatment, starting in 2006. As of 2012, NBC reported that total nitrogen in wastewater inflow to one of their treatment facilities was composed of 61% NH_4^+ and 1% NO_3^- , with the remainder composed of organic N. The final effluent nitrogen composition is 77% NO_3^- , 6% NH_4^+ , and 17% organic N. Moreover,

total N is reduced by 75% (NBC, 2012). During our study period, NBC targeted a monthly average N load of 607 μM N (8.5 ppm) (NBC, 2012). At the beginning of our study, in 2009, NBC used tertiary treatment at one of their plants, and by August 2012, the other plant had come online (achieving their post-upgrade targeted permit level of 5 ppm from May to October). In this study, we focus on the NO_3^- component of WWTF effluent because it is the single largest pool entering the Narragansett Bay ecosystem.

Most of the freshwater input to Narragansett Bay, including both WWTF effluent and river water, enters in the northern reaches through the Providence River and Mount Hope Bay (Fig. 1). The major N inputs to Narragansett Bay have been identified, and ~60% of the total N input is anthropogenic in origin with rivers and wastewater treatment facilities contributing the majority of the anthropogenic N (Nixon et al., 1995, 2008; Krumholz, 2012). As of 2014, tertiary treatment upgrades were in place at many facilities in the Narragansett Bay watershed, including the three largest facilities (Field's Point, Worcester, and Bucklin), with more planned in the near future (Fig. S1). Decreases in N sources to Narragansett Bay were observed for the period of 2006–2010, where wastewater treatment facility N contributions fell by approximately 20% (Krumholz, 2012) due to implementation of tertiary treatment.

To assess the impact of nitrogen to an estuarine system, stable N isotopes (^{15}N : ^{14}N , $\delta^{15}\text{N}$) are often combined with concentration and flux measurements to aid in distinguishing sources (Jordan et al., 1997; Tucker et al., 1999; Costanzo et al., 2001; Cole et al., 2004; Savage, 2005). The $\delta^{15}\text{N}$ value of NO_3^- reflects the $\delta^{15}\text{N}$ value of its source and any transformations to which it was subject. Published values of $\delta^{15}\text{N}$ –

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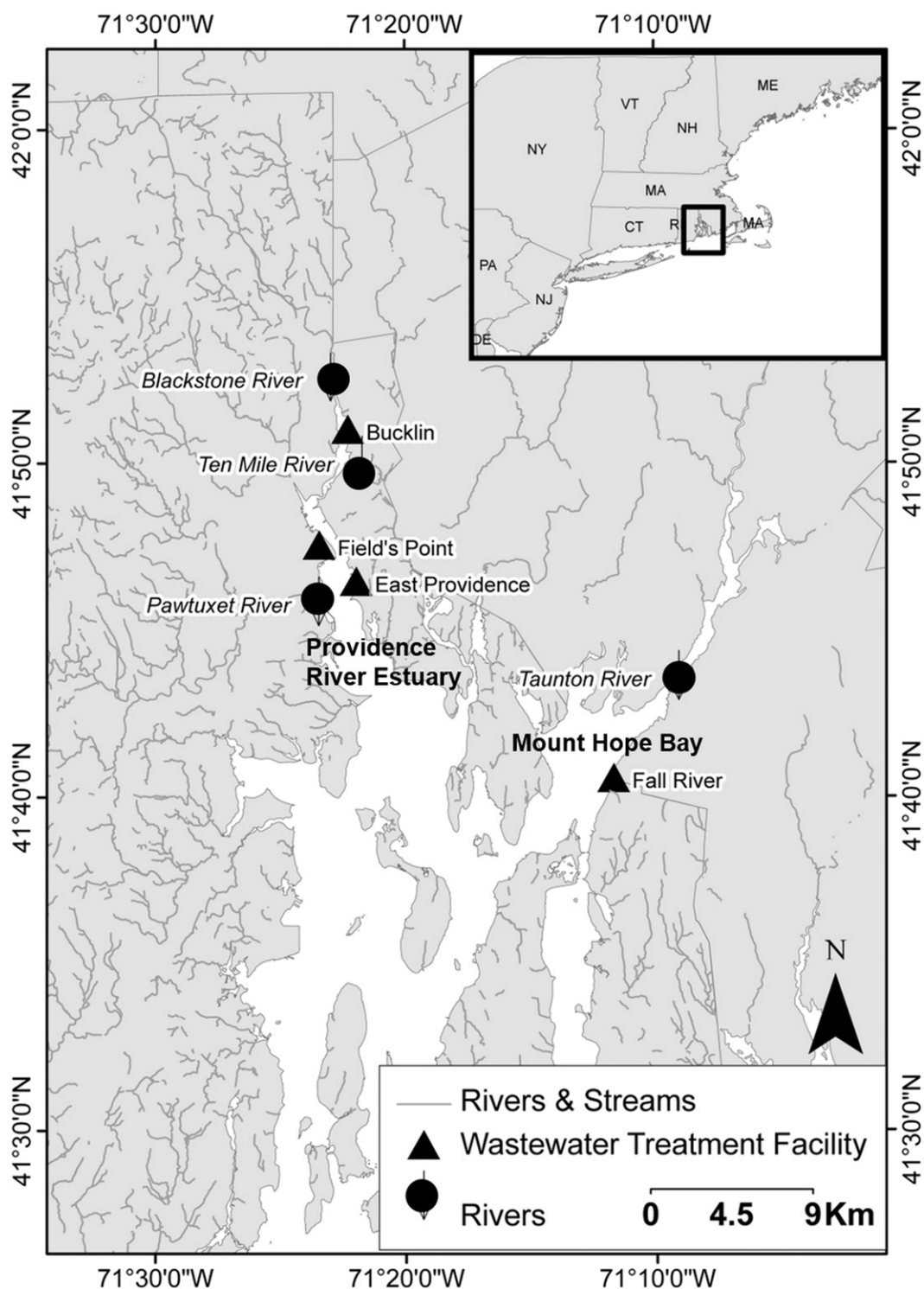


Fig. 1. Narragansett Bay riverine and wastewater treatment facility (WWTF) collection map. Rivers are marked by circles, and WWTFs are marked by triangles. Sampled WWTFs do not discharge into the sampled rivers.

NO_3^- in anthropogenic N from secondary sewage treatment plants, septic system leachate, and rivers range from -3 to $+40\%$ (average $> 10\%$) while marine (near shore) water values range from $+3$ to $+6\%$ (average 5%) (Table S1) (Heaton, 1986; Jordan et al., 1997; Costanzo et al., 2001; Cole et al., 2004; Chaves, 2004; Pardo et al., 2004; Schlacher et al., 2005; DiMilla, 2006; Deutsch et al., 2006, 2009; Dahnke et al., 2008). The overlap in $\delta^{15}\text{N}$ decreases the certainty with which stable N isotopes can be used to uniquely identify anthropogenic discharges. Measurement of the oxygen isotopic composition

($\delta^{18}\text{O}, \%$) of NO_3^- is typically coupled to the $\delta^{15}\text{N}$ measurement, enhancing the ability to distinguish nitrogen sources and nutrient processing pathways (Wassenaar, 1995; Sigman et al., 2001; Mayer et al., 2002; Casciotti et al., 2002; Deutsch et al., 2005; Anisfeld et al., 2007; Saccon et al., 2013; Wexler et al., 2014).

Treatment of wastewater likely imparts variation in $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ values due to fractionation during nitrification (secondary treatment) and denitrification (tertiary treatment). Field work in fresh and salt water demonstrates that fractionation during consumption of nitrate

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