



## Indicators of vehicular emission inputs into semi-arid roadside ecosystems



Julie A. Kenkel <sup>a, \*</sup>, Thomas D. Sisk <sup>a</sup>, Kevin R. Hultine <sup>a, b</sup>, Steven E. Sesnie <sup>a, c</sup>,  
Matthew A. Bowker <sup>d</sup>, Nancy Collins Johnson <sup>a</sup>

<sup>a</sup> School of Earth Sciences and Environmental Sustainability, Northern Arizona University, Flagstaff, AZ 86011-5694, USA

<sup>b</sup> Department of Research, Conservation and Collections, Desert Botanical Garden, Phoenix, AZ 85008, USA

<sup>c</sup> U.S. Fish and Wildlife, Southwest Natural Resources Inventory and Monitoring Program, Southwest Region, Albuquerque, NM 87103-1306, USA

<sup>d</sup> School of Forestry, Northern Arizona University, Flagstaff, AZ 86011-15018, USA

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### ABSTRACT

The relative importance of pollution from non-point sources, such as vehicular emissions, is not well understood in sensitive semi-arid protected areas, such as Grand Canyon National Park (GCNP). Roadside N deposition could be substantial in areas with high visitation and traffic volume such as GCNP, although inputs of roadside pollution have not been quantified in GCNP. We used three potential indicators of N enrichment to evaluate impacts of vehicle emissions on roadside ecosystems in GCNP: 1) concentration of nitrogen oxides (NO<sub>x</sub>) captured by passive air samplers, 2) natural  $\delta^{15}\text{N}$  abundance in the foliage of mature Piñon pine trees and soil N, and 3) concentrations of available inorganic N (NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>) in soil. Over an eight-month period, N enrichment was assessed at ten sites located across hypothesized gradients of vehicular N deposition based on distance from primary roadways and traffic levels in GCNP. Ogawa passive air samplers indicated that NO<sub>x</sub> levels were on average 52% higher at roadside locations compared to 30 m away, and were highest at the most heavily trafficked sites in the park. Natural abundance  $\delta^{15}\text{N}$  of foliage was significantly higher ( $F = 16.04$ ,  $p = 0.007$ ,  $n = 237$ ) at roadside locations than at 15, and 30 m distances: a pattern that is consistent with local N inputs from vehicle emissions. Available inorganic soil N was significantly higher ( $F = 3.46$ ,  $p = 0.013$ ,  $n = 8$ ) at the South Entrance roadside compared to areas with lower traffic densities. The direct measurement of atmospheric NO<sub>x</sub> using Ogawa samplers and  $\delta^{15}\text{N}$  of Piñon pine needle tissues were the best indicators of roadside N deposition. Our results also suggest that vehicular emissions on the South Rim of GCNP were incorporated in the plant nutrient pools near roadside environments. Although N emissions in GCNP comply with current annual air quality standards as specified by the Clean Air Act, it remains unclear whether the standards are low enough to sustain ecological integrity of native ecological communities along roadsides. Coupled with information on N-pollution tipping points and other ecological indicators, the metrics used here may prove to be a cost effective approach for monitoring roadside N-deposition and informing management decisions.

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### 1. Introduction

Anthropogenic enrichment of biologically reactive forms of nitrogen (N) changes nutrient regimes in soils, alters plant community composition, and reduces biological diversity (Vitousek et al., 1997; Bobbink et al., 2003; Bowman et al., 2006; Báez et al.,

2007). Because N is often the most limiting nutrient in terrestrial systems, even small levels of N enrichment may have measurable impacts on plant and soil communities unaccustomed to additional N inputs (Vitousek, 1982; Fenn et al., 2003). Semi-arid ecosystems may be particularly sensitive to N deposition, although the extent of ecological impacts is unknown (Belnap and Harper, 1990; Padgett et al., 1999; Baron et al., 2000).

Levels of atmospheric N deposition and associated ecosystem impacts in the western U.S. continue to increase. Although

\* Corresponding author.

E-mail address: [jkenkel@gmail.com](mailto:jkenkel@gmail.com) (J.A. Kenkel).

atmospheric N deposition in the western U.S. is historically lower than other regions, studies throughout the western states report deleterious ecosystem shifts attributable to increased N deposition through time (Weiss, 1999; Baron et al., 2000; Fenn et al., 2003, 2008; Rao et al., 2010; Reed et al., 2013; Ellis et al., 2013). Ecosystem responses to increased N deposition in the arid and semi-arid southwestern U.S. differ from those in more mesic regions because productivity is primarily limited by water, secondarily by N, and is pulse-driven (Noy-Meir, 1973; Romney et al., 1978; Gutierrez et al., 1988, 1992; Collins et al., 2010). Although N deposition in semi-arid and arid regions is generally lower than in mesic regions, it represents a 6–9 fold increase since the beginning of the industrial revolution, and will continue to increase with population growth (Collins et al., 2010). The impacts of anthropogenic N enrichment in highly sensitive arid regions with characteristic limitation of both water and N are not well understood. Therefore, quantifying these impacts is critical for understanding ecological responses to projected changes associated with growth of human populations in semi-arid regions.

Grand Canyon National Park (GCNP) in northern Arizona is considered a class 1 area, meaning that the park must strive to meet the cleanest air quality conditions. To meet these goals, GCNP monitors both regional and local air pollutants using atmospheric deposition models and monitoring programs. For example, the Comprehensive Air Quality Model with extensions (CAMx), the National Atmospheric Deposition Program (NADP), and the Clean Air Status and Trends Network (CASTNet) monitor atmospheric N deposition trends from point and non-point emission sources. These programs detect N deposition in GCNP to range between 1 and 4 kg ha<sup>-1</sup> yr<sup>-1</sup> (<http://java.epa.gov/castnet/>, <http://nadp.sws.uiuc.edu/NTN/annualmapsbyyear.aspx>), which is within the recent critical N load limit recommended for GCNP (2.5–5 kg ha<sup>-1</sup> yr<sup>-1</sup>; Blett et al., 2014). However, both modeling and monitoring assessments of N deposition may underestimate total N deposition because they monitor only select species of N, are located in rural areas away from vehicular traffic, and operate at an inadequate resolution to capture deposition dynamics on small scales within park management jurisdiction, such as roadside gradients (Baumgardner et al., 2002; Fenn et al., 2003; Redling et al., 2013).

Grand Canyon National Park is ranked as a highly sensitive ecosystem and of high management priority for National Park Service; however, it is challenged to meet its resource protection aims due to air pollution from local and regional sources both within and outside of park jurisdiction (National Park Service, 2011). The National Park Service recognizes anthropogenic N deposition as a threat to natural resource conservation and its organizational mission (National Park Service, 2011; Blett et al., 2014). Long-distance transport of pollution from large metropolitan areas and point sources such as coal-fired power plants are known to reduce air quality in GCNP (Diem, 2007; Terhorst and Berkman, 2010). Additionally, GCNP is the second most visited park in the U.S.; each year, over 4 million tourists visit the park, and 97% of these visitors tour the park in their personal vehicles (National Park Service, 2011; Cui et al., 2013).

Roadside pollution may cause deleterious changes in vegetation (Gratani et al., 2000; Viskari et al., 2000; Honour et al., 2009). Plants take up nutrients not only through roots, but can also access atmospheric NO<sub>x</sub>, HNO<sub>3</sub>, and NH<sub>3</sub> through stomata (Wellburn, 1990; Thoene et al., 1991; Garten, 1993; Padgett et al., 2009). Nitrogen oxides (NO<sub>x</sub>) from tailpipe emissions of automobiles and other vehicles are known to have a fertilization effect on heathland, grassland, and non-vascular vegetation along roadsides (Angold, 1997; Weiss, 1999; Pearson et al., 2000; Gelbard and Belnap, 2003). Plants growing in mesic environments with elevated N

levels show increased susceptibility to pathogens and other biotic (including invasive species) and abiotic stressors (Bobbink, 1991; Nordin et al., 1998; Power et al., 1998; Steers and Allen, 2010). Riddell et al. (2008) found that under lab-simulated dry and humid conditions, the lichen species *Ramalina menziesii* collected from “pristine conditions” showed significant reductions in chlorophyll content and carbon exchange capacity when exposed to nitric acid (HNO<sub>3</sub>) gas, a secondary pollutant created through reactions between water and nitrogen. However, less is known about the impacts of roadside pollutants, such as (NO<sub>x</sub>), on roadside vegetation in drylands where vegetation may respond to concomitant water limitation and N deposition from vehicular emissions (Forman et al., 2003). Soil N concentration is highest when water availability is low; however, without water, plants and microbes are relatively inactive and unable to use the additional N as a resource (Cregger et al., 2014). Therefore, the pulses of precipitation typically experienced in drylands may cause nutrient uptake events; where otherwise plants/microbes cannot metabolize nutrients from atmospheric deposition unless coupled with water events. Semi-arid drylands present different environmental challenges than mesic areas, requiring further investigation to understand nutrient cycling regimes.

Natural fluctuations and assimilation of N in vegetation and soils make it difficult to assess anthropogenic N inputs; however, the analysis of stable isotope ratios offers a method to trace pollution to its source. Because different sources of N often have distinctive isotopic ratios (i.e. relative abundance  $\delta^{15}\text{N}$ ), N pools in the atmosphere, plant tissues and soils can often be traced back to their sources (Kendall et al., 2007). For example, the  $\delta^{15}\text{N}$  of NO<sub>x</sub> derived from fuel combustion in vehicles with catalytic converters range from +3.7‰ to +5.7‰ (Moore, 1977; Ammann et al., 1999; Pearson et al., 2000; Middlecamp and Elliott, 2009), while vehicles without catalytic converters range from –13‰ to –2‰ (Heaton, 1990). Natural NO<sub>x</sub> sources, including lightning (–0.5–1.4‰; Hoering, 1957) and biogenic N<sub>2</sub>O emissions from fertilized soils have lower  $\delta^{15}\text{N}$  values (–49‰ to –20‰; Hoering, 1957; Li and Wang, 2008). The  $\delta^{15}\text{N}$  signatures of plants along roadsides are often elevated compared to the surrounding environment (Pearson et al., 2000). In this way,  $\delta^{15}\text{N}$  analysis can be used to track the origin of NO<sub>x</sub> in the atmosphere as well as the sources of N in soils and plants.

Spatial variability in the distribution of atmospheric N deposition is not well known in the Grand Canyon Region, and may be of increasing importance to guide park management decisions. High levels of tourism, known vehicular patterns, and the sensitivity of GCNP make it an ideal location to investigate indicators of N-deposition from vehicular emissions in a primarily water-limited environment. In this study, we combined measurements of localized atmospheric NO<sub>x</sub> concentrations, foliar and soil  $\delta^{15}\text{N}$  abundance and soil N concentrations to evaluate N enrichment along roadsides in GCNP. In this study, we combined measurements of localized atmospheric NO<sub>x</sub> concentrations, foliar and soil  $\delta^{15}\text{N}$  abundance and soil N concentrations to evaluate N enrichment along roadsides in GCNP. Areas of GCNP closest to the road at the South Entrance with were predicted to receive the highest N inputs, particularly during the peak tourist season.

## 2. Materials and methods

### 2.1. Study site

The study was conducted from May 2011 to January 2012 at ten sites along the South Rim of Grand Canyon National Park in northern Arizona (Fig. 1a). There are traffic density gradients in GCNP. Nearly 85% of approximately 2.5 million vehicles enter the

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