Increased vulnerability of urban areas from atmospheric N deposition

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Atmospheric emissions of reactive nitrogen (N) are a serious problem for ecosystem and human health, having implications for both air and water quality. Nitrogen atmospheric deposition (AD) in urban areas is primarily composed of nitrate (i.e. nitric acid (HNO_3), or particulate NO_3^-) and ammonium (NH_4^+). NO_3^- is the primary sink of N oxides from the atmosphere, while NH_4^+ is a sink for gaseous NH_3. Until recently, NO_3^- was the dominant N component of precipitation and particulate matter. Due to significant decreases in N oxides emissions through control policies, NO_3^- deposition has significantly decreased. However, NH_4^+ deposition has continued to increase and now dominates AD. The U.S. National Atmospheric Deposition Program (NADP) monitoring locations are specifically located to assess well-mixed background concentrations, and tend to be located in rural areas. Recent observations and atmospheric chemistry modeling, suggest that significant concentration gradients in AD exist between rural and urban areas. Thus, the environmental impact of AD, especially on water quality near urban areas, may be significantly underestimated or unknown.

To better quantify AD in an urban region and understand the impact on coastal biogeochemistry, event-based precipitation was collected year-round in Providence, RI. Inorganic N concentrations (NH_4^+ and NO_3^-) were quantified and compared with the nearest NADP monitoring locations, including an urban site (Boston, MA), rural site (Abington, CT), and an oceanic/coastal site (Cape Cod, MA). We find that urban sites have higher N concentrations than rural and oceanic sites for all months of the year. Interestingly, the event-based urban data shows significant enhancements compared to the surrounding sites, with as much as 8x higher NH_4^+ concentrations (maximum $[NH_4^+]$ = 352.5 μM) and 4x higher NO_3^- concentrations (maximum $[NO_3^-]$ = 292.0 μM). This suggests that urban deposition has been largely underestimated compared to well-mixed monitoring sites, and implicates the influence of an urban footprint. Moreover, the NH_4^+/NO_3^- ratio is higher in rural and urban sites compared to the oceanic site, suggesting urban sources of NH_3 are important for N deposition. The isotopic compositions of NO_3^- and NH_4^+, along with analysis of atmospheric transport pathways for each event, will be used to constrain local versus transported precursor emission sources. The influence of different sources on the composition of precipitation will also be assessed, using recent well-constrained isotopic measurements of precursor gaseous species from vehicles, agricultural soils, and wildfire plumes in the U.S.