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Atmospheric Nitrogen Deposition: Analysis of a little studied, but important, piece of the N budget in Narragansett Bay

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INTRODUCTION

Atmospheric deposition is categorized as a substance, mostly chemical, which falls to the ground. Atmospheric deposition can derive from natural as well as anthropogenic sources. Natural sources include forest fires, lightning, volcanic eruptions or outgassing, and emissions from soils while anthropogenic sources include factories and cars (incomplete combustion) or large-scale agricultural practices (Cornell et al. 1995; Cornell et al. 2003; Howarth et al. 1996, 2002; Howarth and Rielinger 2003). The particles are swept up into the atmosphere and transported (either locally or long-distance) to other areas. During transport, chemical or physical changes may occur. The chemicals are deposited to the landscape as wet or dry deposition. Wet deposition occurs during precipitation, while dry deposition occurs when particles and aerosols settle out of the air. Both types of deposition can have a direct or indirect effect on a water body. Direct deposition is that which falls on the water body itself, while indirect deposition is that which falls on the land, and is transported to the water body of interest through run-off or riverine discharge (Howarth et al. 1996, 2002; Howarth and Rielinger 2003).

Until a breakthrough study by Fisher and Oppenheimer (1991), atmospheric nitrogen (N) deposition was little studied or underestimated. They and others since have found that N deposition is mainly caused by fossil fuels combustions and ammonia volatization from agricultural sources with short atmospheric lifetimes (hours to days) which keep N compounds near their sources (Fisher and Oppenheimer 1991; Paerl 1993; Cornell et al. 1995; Howarth et al. 2002; Gao et al 2007). Organic N can account for ~20-65 % (approximately 38% in North America) of total dissolved N in precipitation (Zhang and Anastasio 2001; Weathers et al. 2000). Direct deposition of N, as either dissolved inorganic N (DIN as nitrate (NO_3^-), and ammonium

(NH_4^+)) or dissolved organic N (DON), accounts for 1-40 % of total N inputs to estuaries (Nixon et al. 1996; Paerl 1997; Paerl and Whitall 1999; Valigura et al 2000; Howarth 2008). Direct deposition is most significant in large systems where there is a lot of surface area to collect the deposition (Nixon et al. 1996), and in coastal systems with small watersheds in comparison to the area of surface waters (Zarbock et al. 1996). Indirect deposition, rather than direct deposition, is considered the major route of N input to estuaries. Five to 80% of the total N flux in major rivers of New England could be accounted for by measuring indirect deposition and combining with direct and dry deposition or through use of the SPARROW model (Alexander et al. 2001; Boyer et al. 2002; Howarth and Rielinger 2003; Howarth 2008). The wide range comes from uncertainty in dry and indirect deposition measurements or estimations. Most measurements are of direct, wet deposition, with indirect deposition numbers estimated, and few dry deposition studies. Most dry deposition numbers are estimated as part of national programs (CASTNet and AIRMON-Dry). CASTNet operates one site in southern New England, in Abington, CT, far away from nitrate emission sources (http://epa.gov/castnet/javaweb/index.html).

In Narragansett Bay, direct deposition of N accounts for about 6 % of the total N budget to the Bay, with the majority of N derived from sewage (Nixon et al. 1995, 2008; Krumholz 2012). Indirectly deposited N comes through storm water runoff and riverine sources. Forested regions export over 20% of the N deposited on them through rivers, and potentially export more N when the system's nutrient needs are met (van Breeman et al. 2002). Boyer et al. (2002) calculated that atmospheric deposition accounts for one-third of the N in the Blackstone River watershed (a major tributary of Narragansett Bay). Using this estimate, Howarth 2008 calculated that the

combination of direct and indirect deposited N would make up about 30 % of the total N input to the Bay. While N input derived from wastewater treatment facilities is still larger, atmospheric deposition should be a monitoring and management focus.

Atmospheric deposition continues to be an overlooked part of the Narragansett Bay N budget. The last published study on atmospheric N deposition was in 1991, using data collected in 1988-1990. Since then, data were collected from 1992-1996, but collection fell off after that period. The goals of this paper are to introduce the 1992-1996 wet atmospheric DIN and DON data, and compare the data to previously collected atmospheric deposition data from Narragansett Bay, the northeast US, and the National Atmospheric Deposition Program (NADP).

METHODS

Site Description

Narragansett Bay, including Mount Hope and Greenwich Bays and the Providence River Estuary, is 328 km^2 and has a mean depth of 8.3 m (Fig. 1) (Pilson 1985). Freshwater input is relatively low (around 100 m^3s^{-1}) and most of the input occurs in the urbanized northern reaches, near the city of Providence, where most of the watershed's population lives (Nixon et al. 1995; Krumholz 2012) (Fig. 1). Circulation is tidally driven; with ocean water entering the bay through the East Passage, and out the West Passage. Salinity is high throughout the estuary (between 20 to 32 ppt) with the mouth of the bay the most saline (about 32 ppt).

From November 1992 through December 1996, rain water samples were collected from two sites: Prudence Island, in the Narragansett Bay National Estuary Research Reserve (NBNERR)

and the Graduate School of Oceanography (GSO) (Fig. 1). Prudence Island is further north than GSO, in the heart of Narragansett Bay. GSO is located on the shores of the lower Bay, in Narragansett, RI and is the oceanography school of the University of Rhode Island. The two sites are located, at their closest, about 7 miles apart.

Field Collection

Samples were collected by placing acid-washed PVC funnels into acid-washed bottles. The bottles were placed on level surfaces, clear of any overhang from buildings or trees. After each rainfall, the bottles were collected, brought to the lab, filtered, and frozen until analysis.

Laboratory Analysis & Calculations

All samples were analyzed for nutrients on a Lachat QuickChem 2000 flow injection autoanalyzer using standard colormetric methods for NO_{3+2}, and NO_2^- using EPA method 353.4 (Grasshoff 1976; US EPA 1997) in the Nixon laboratory at the University of Rhode Island (URI) (minimum detection $[NO_3^-] = 0.02$ μM). Statistical analyses were performed to establish any significant difference between the sites. Any precipitation data needed for flux analysis was obtained through NOAA's National Climate Data Center, for T.F. Green Airport, located in Warwick, RI (http://www.ncdc.noaa.gov). Flux calculations were made by multiplying the concentration (in μmol m^{-3}) by the rainfall volume (in mm). Flux units are μmol m^{-2} rain $event^{-1}$ or mmol m^{-2} rain $event^{-1}$. Yearly averages are in mmol N m^{-2} y^{-1}.

RESULTS & DISCUSSION

1992-1996 Data

The largest component of atmospheric deposition in Narragansett Bay was wet deposition of nitrate (NO_3^- + NO_2^-; referred to hereafter as NO_3^-) (average of 1.9 mmol N m^{-2} y^{-1}). Ammonium (NH_4^+) contributed 1.1 mmol N m^{-2} y^{-1}, on average, while dissolved organic nitrogen (DON) contributed 0.7 mmol N m^{-2} y^{-1}, on average. The yearly average of dissolved inorganic nitrogen (DIN, NO_3^- plus NH_4^+) wet deposition was 3.0 mmol N m^{-2}, and total N deposition was 3.7 mmol N m^{-2} y^{-1}. During the year, fluxes were not equal in magnitude. Nitrate had the largest magnitude (< 6 mmol m^{-2} rain $event^{-1}$), followed by ammonium (< 2 mmol m^{-2} rain event), and DON (< 1.5 mmol m^{-2} rain $event^{-1}$).

In his thesis, Fraher tested whether the flux of N was more dependent on rainfall amount or concentration of the flux, finding that the flux of N was more dependent on rainfall (Fraher 1991). The same analysis found that from 1992 to 1996, flux was dependent on rainfall amount for nitrate NO_3^-, NH_4^+, and dissolved organic nitrogen (DON) (Fig. 2), in support of Fraher's findings (Fraher 1991). In addition, an unpaired t-test was used to determine if the data from both GSO and Prudence Island can be combined into one Narragansett Bay dataset. Data from GSO and Prudence Island were not statistically different for NO_3^- or NH_4^+ and were treated as one data set ($p = 0.95$ and $p = 0.90$, respectively). DON samples appeared to be statistically different between both sites ($p < 0.0001$), and the GSO averaged (191 μmol m^{-2} rain $event^{-1}$) higher than Prudence Island (86 μmol m^{-2} rain $event^{-1}$). Therefore, the data were not combined. The difference in [DON] between sites may be due to their geography. More DON is thought to deposit on land rather than the ocean (Russell et al. 1998; Cornell et al. 2003). The GSO site was

mostly surrounded by land, while the Prudence Island site was mostly surrounded by water. Therefore the GSO site may have more DON input than the Prudence Island site.

Seasonally, the largest fluxes for NH_4^+, NO_3^- and DON appear to be in the summer. Over the time period, the flux for all parameters did not change (Figs. 3-5). For nitrate and ammonium, however, a small, though not statistically significant, increase in flux was noted between spring and summer, with a decrease through fall (Figs. 3-5).

Comparison with other Narragansett Bay data

Fraher's 1991 thesis contained the first comprehensive analysis of atmospheric DIN deposition in Narragansett Bay. His work has been used as the atmospheric deposition portion of the Narragansett Bay N budget at least since Nixon et al. 1995. Fraher did not collect any DON data. However, from 1985 to 1986, Nowicki and Oviatt (1990) collected atmospheric deposition data during a unrelated experiment. The current data will be compared with theirs.

Rainfall was first analyzed to compare the 1992-1996 dataset to Fraher's because deposition is highly dependent on rainfall (Fig. 2) and if rainfall has changed throughout the eight year period, deposition may have changed as well. Rain did not changed between 1998 and 1996 ($p = 0.7$; Fig. 6). Multiple studies on Narragansett Bay (Pilson 2008; Smith et al. 2010) showed that rainfall has increased over the last half century or so. During the sample period for which we have atmospheric deposition data, rain remained stable across the years (Fig. 6). No seasonal or monthly changes were noted, either. Long-term changes in rainfall could alter the amount of wet

deposition to a region. However, with this limited data set, we cannot say how the increase in rainfall would impact N deposition to Narragansett Bay.

Atmospheric deposition of DIN did not change between 1988 and 1996 ($p = 0.06$; Figs. 3-5). A quick glance at the 1988-1990 and the 1992-1996 data sets shows that DIN deposition appeared to be greater from 1992-1996. Statistically, this is not the case ($p = 0.06$), and the differences may be related to how the samples were collected. From 1988-1990, Fraher used an automated collector at his stations. This collector opened and closed upon sensing the start and end of the storm. The 1992-1996 data were collected by placing a funnel inside a bottle in an open area before a storm and collected the bottle after. A strong possibility exists that some dry deposition was collected with the rainfall, artificially inflating the concentration of DIN in some of the samples.

During 1985-1986, Nowicki and Oviatt (1990) collected atmospheric DON data during a nutrient enrichment experiment in the Marine Ecosystems Research Lab's mesocosm tanks. During their study, they found that DON contributed 2.8 – 35.7 μmol m^{-2} rain $event^{-1}$. From 1992-1996, DON ranged from 1 – 1426 μmol m^{-2} rain $event^{-1}$ (averages: 191 and 86 μmol m^{-2} rain $event^{-1}$ for GSO and Prudence Island, respectively). The differences between the studies could be from collection technique, amount of sampling, or uncertainties within the measurement. Both studies had similar collection techniques (funnels fitted in the mouths of acid-washed bottles, left out during the precipitation event). However, the current dataset collected precipitation from more events than the Nowicki and Oviatt (1990) study, and therefore, probably captured more seasonal variation, and potentially stronger storms, which would bring more DON. Additionally,

measuring DON with confidence has been a challenge (Cornell et al. 2003). Usually, DON is measured by subtracting the inorganic N concentration from the total N. Uncertainties within measuring total N and inorganic N will compound, creating more uncertainty for the organic N measurement (Cornell et al. 2003). Nowicki and Oviatt (1990) also found that DON and NO_3^- deposition were strongly correlated, but this was not replicated in the current study at either GSO or Prudence Island.

Comparison with regional and national trends

Seasonally, both Narragansett Bay data sets (Fraher, 1991, and 1992-1996) compare well with other stations in the Northeast (Fig. 7), including Long Island Sound, and Lewes, DE. Both these sites are part of the National Atmospheric Deposition Program (NADP). However, the Narragansett Bay deposition data is less than Sandy Hook and Tuckerton (Fig. 7). Gao et al. (2007) attributes the high amount of N deposition at this site to regional anthropogenic influence from the New York and Philadelphia metropolitan areas.

Nationally, the Northeast deposition data do follow NADP trends (Fig. 8). The Long Island Sound station appears to be lower than the other stations. This could be from a more direct oceanic influence, whereas the other stations may be influenced by westerly winds in the summer. Sandy Hook is once again higher than the national trends, which could be from a localized hot-spot of deposition.

DON is comprised of a diversity of organic materials, from both anthropogenic and natural sources. Most DON is deposited on land, even though it is a more significant component of total

N deposition to marine environments (Russell et al. 1998; Cornell et al. 2003). Russell et al. (1998) measured DON flux in Cheasapeake Bay at 0.17 mmol m^{-2} d^{-1}, which equals 62 mmol m^{-2} y^{-1}, which is much higher than the current Narragansett Bay measurements. Regional differences could be significant and come from the source of nitrogen itself, whether more factory driven or agriculturally driven.

CONCLUSIONS

Fraher (1991) established a baseline for N deposition research in Narragansett Bay, which was extended between 1992-1996. However, further research is needed with longer datasets to establish long-term trends. Multiple studies (Pilson 2008 and Smith et al. 2010) have shown that rainfall increased by 30 cm total from 1895 to 1999 in Narragansett Bay. Will deposition change as well? Without a long term dataset, we won't know the answer. This information is also vitally important to nitrogen budgets for the bay. With decreases from anthropogenic sources of N, such as wastewater treatment facilities, atmospheric deposition may because a relatively larger piece of the budget. While direct deposition makes up about 6% of the budget (Nixon et al. 1995, 2008; Krumholz 2012), Howarth (2008) calculated that indirect plus direct deposition makes up about 30% of the N budget. Decreases to other point-source N inputs may make atmospheric deposition relatively more important. Using the 2010 N budget (Krumholz 2012), the current amount of N atmospheric deposition is about 4%. While this is a slight decrease in direct deposition, we still have very little knowledge of how deposition changes over longer time periods. Therefore, we need longer-term monitoring of this little studied piece of Narragansett Bay's nitrogen budget.

MANAGEMENT IMPLICATIONS

Atmospheric deposition is an often-overlooked portion of the N cycle in urban estuaries, especially in regions like Narragansett Bay, where atmospheric deposition makes up about 5% of the N budget (Nixon *et al.* 2008). However, Narragansett Bay has enacted pollution policies aimed at reducing eutrophication effects of anthropogenic N. These policies would reduce point source N loading to the bay by about 35% and have already accounted for a 20% reduction in N inputs to the Bay (Nixon et al. 2008, Krumholz 2012). This reduction could make other sources of N, like atmospheric deposition, more important. In light of that increased importance, continued monitoring is prudent.

The National Atmospheric Deposition Program is a national program which monitors precipitation chemistry. It began in 1977, and since then has increased to include the National Acid Precipitation Assessment Program, the Atmospheric Integrated Research Monitoring Network, the Mercury Deposition Network, and the Atmospheric Mercury Network. Over 30 years, this program has supplied data from every state, with the exception of Rhode Island (http://nadp.sws.uiuc.edu/). Connecticut, New York, and Massachusetts have sites within the NADP and geographically close to Rhode Island, however those sites are rural in nature (either in Northeastern CT, on the North Shore of Long Island, or Eastern MA, on Cape Cod; http://nadp.sws.uiuc.edu/sites/ntnmap.asp).

Rhode Island has a very unique estuary when compared to its neighbors (Cape Cod, MA, Northeastern CT, and Long Island, NY). The head of the bay is highly urbanized, while the mouth of the bay is fairly suburban. While the regional NADP sites are useful to get regional

trends (Figs. 7 and 8), they cannot be used in Narragansett Bay N budgets. According to the most comprehensive atmospheric deposition project completed in Narragansett Bay to date, using rural Massachusetts stations as a proxy for urban Narragansett Bay atmospheric deposition would underestimate nitrogen deposition by 30-50 % (Fraher 1992). To this end, it is extremely important that monitoring continue in Rhode Island. Not only would RI have its own NADP site, the data gathered can be used to accurately account for atmospheric deposition in nutrient budgets.

Monitoring Program Needs

The analysis of atmospheric deposition data collected on Prudence Island from 1992 to 1996 led to realization that Rhode Island does not, currently, have a long-term monitoring site dedicated to collecting deposition data. As a final part of this analysis, I researched the possibility of long-term site placement on Prudence Island by contacting the National Atmospheric Deposition Program.

The NADP would like that the group running the site have the data open to everyone. Rhode Island does not already have a station, so the NADP would be very happy to have coverage here. For a successful application, the most important criteria are that the site sits in an open area, and the monitoring group (NBNERR) follows the standard operating procedures set up by NADP. The site needs to have 110v power (though a solar powered collector is accepted). Finally, the site can contract with the federal government, so NBNERR, as part of a federal agency, will be allowed to conduct the monitoring. In addition to the mechanical qualifications, dedicated

personnel need to visit the collector every Tuesday between 8 and 10 am to collect the sample bottle and clean the machine. This will take about 15 minutes.

The start-up will cost about $10,000 for the rain gauge and precipitation collector (NBNERR will have a choice of two), with an average carrying cost of $6,000 per year. The yearly cost includes weekly samples, all lab analysis costs, quality assurance and control, plus upkeep of the data on a central data management system. The data will include rain amount, pH, conductivity, NO_3^-, NH_4^+, PO_4^{3-}, and other cation concentrations. The reserve will need to provide a dedicated person to attend the collector weekly to collect the bottle and maintain the machine, about 15 minutes of time per week.

In fiscally conservative times, finding the start-up and maintenance costs could be problematic. A solution to this would be to coordinate with other local agencies and universities to subsidize some or all of the costs. Potential collaborations include University of Rhode Island, Brown University, and Roger Williams University.

If start-up and maintenance funding, and personnel can be procured, this is a valuable opportunity for NBNERR. The nearest atmospheric deposition monitoring stations are near Hartford, CT and on Cape Cod, MA. These sites do not represent Narragansett Bay in terms of land use percentages and population density. The only way to truly understand atmospheric deposition dynamics of Narragansett Bay is to have a permanent monitoring station. The NBNERR on Prudence Island meets all the requirements, and is uniquely centered in the bay, making it the perfect place.

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Figures

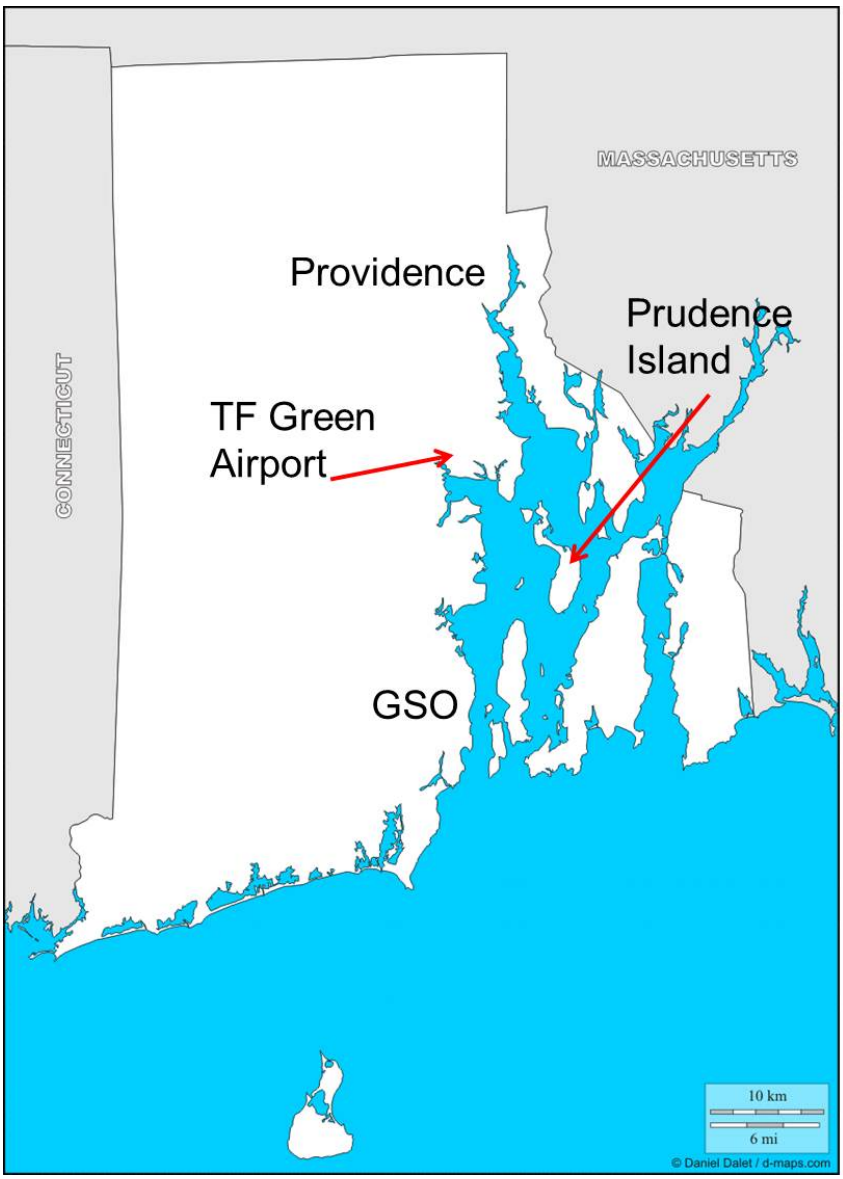

Figure 1. Narragansett Bay atmospheric deposition collection map. Sample sites are labeled (Prudence Island and GSO), as is the city of Providence, and T.F. Green Airport, the site of the precipitation data collection.

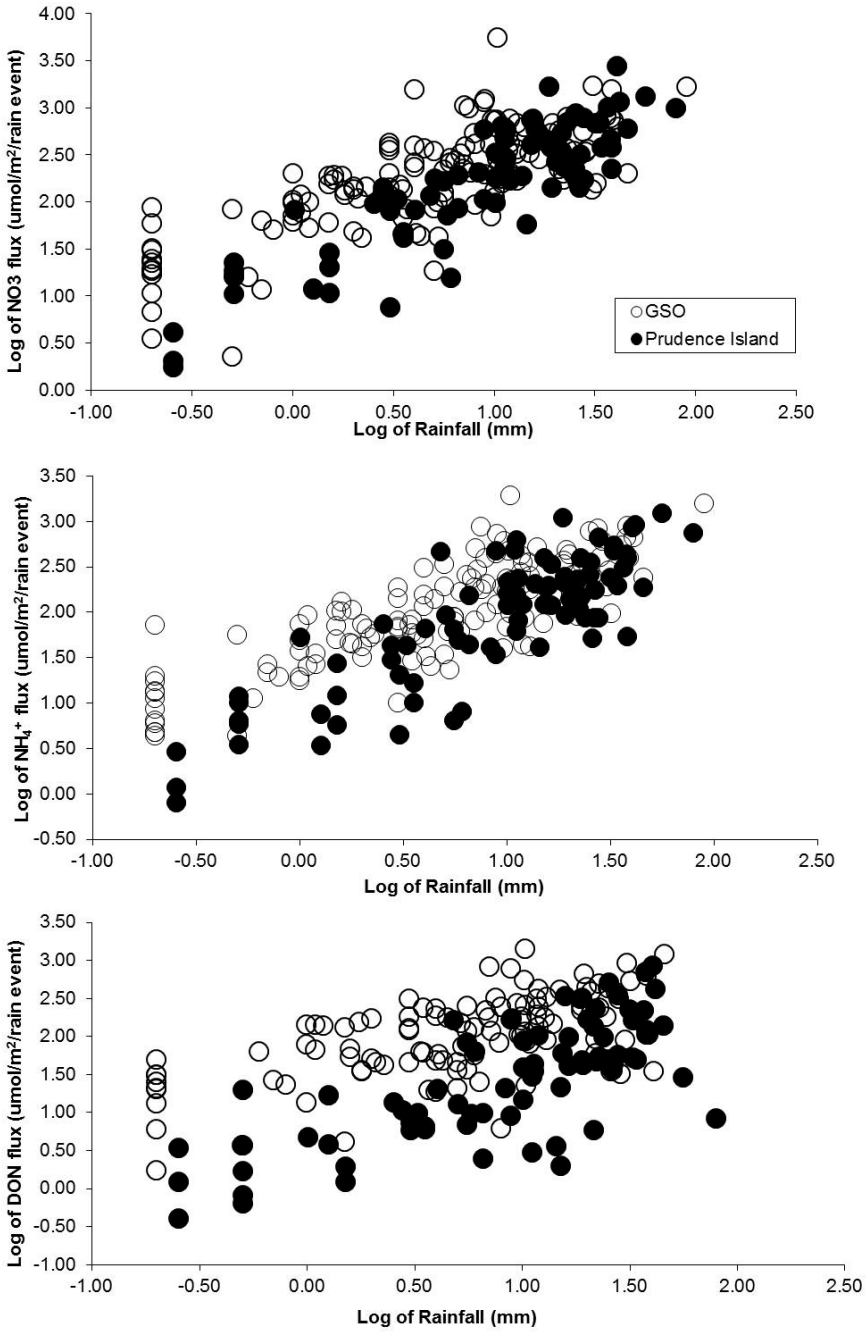

Figure 2. Flux of NO_3^- (top panel), NH_4^+ (middle panel), and DON (bottom panel) versus rainfall. All values have been log-base 10 transformed for statistical purposes. Flux is in units of umol/m^2/rain event, and rainfall is in mm.

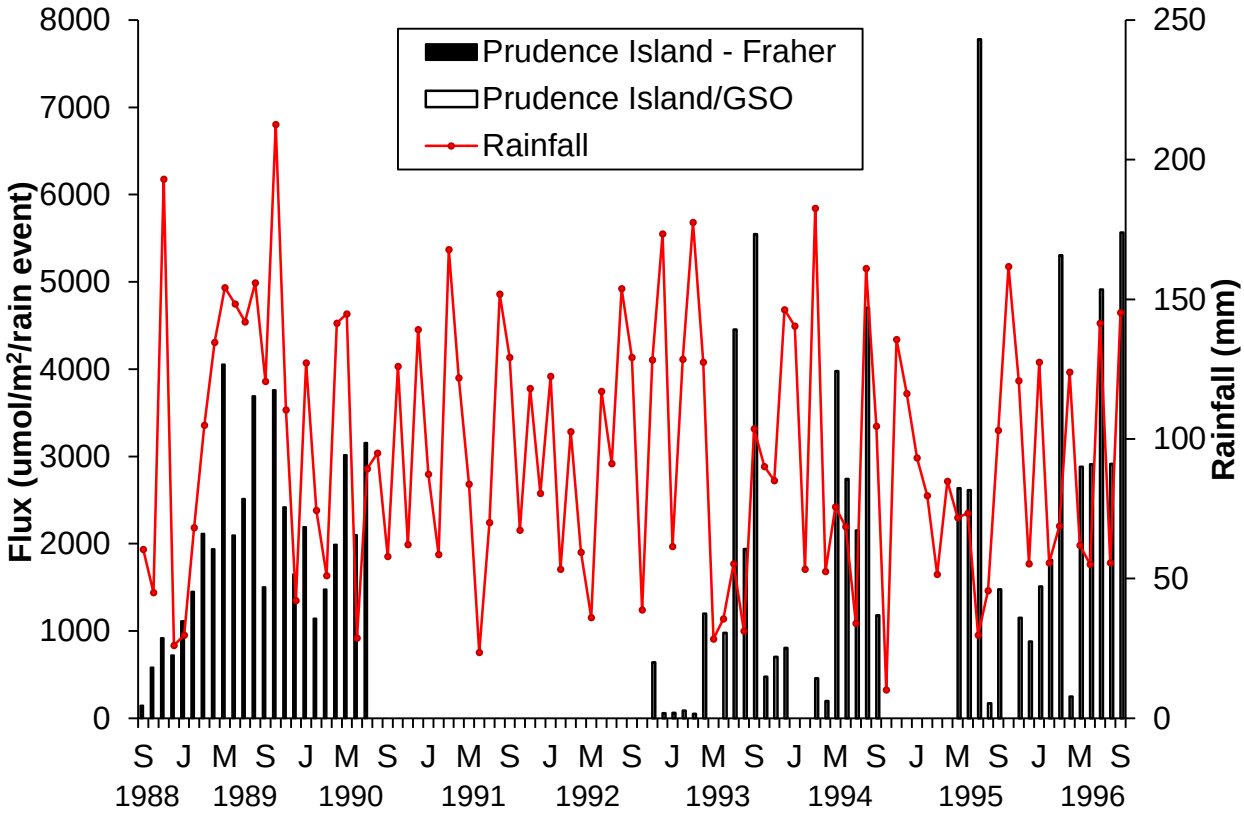

Figure 3. Flux of NO_3^- versus month (S = September, J = January, and M = May) and year. Vertical bars represent data from 1988-1990 (left; Fraher 1991), and data from 1992-1996 (right), while the red line represents monthly rainfall totals from 1988-1996.

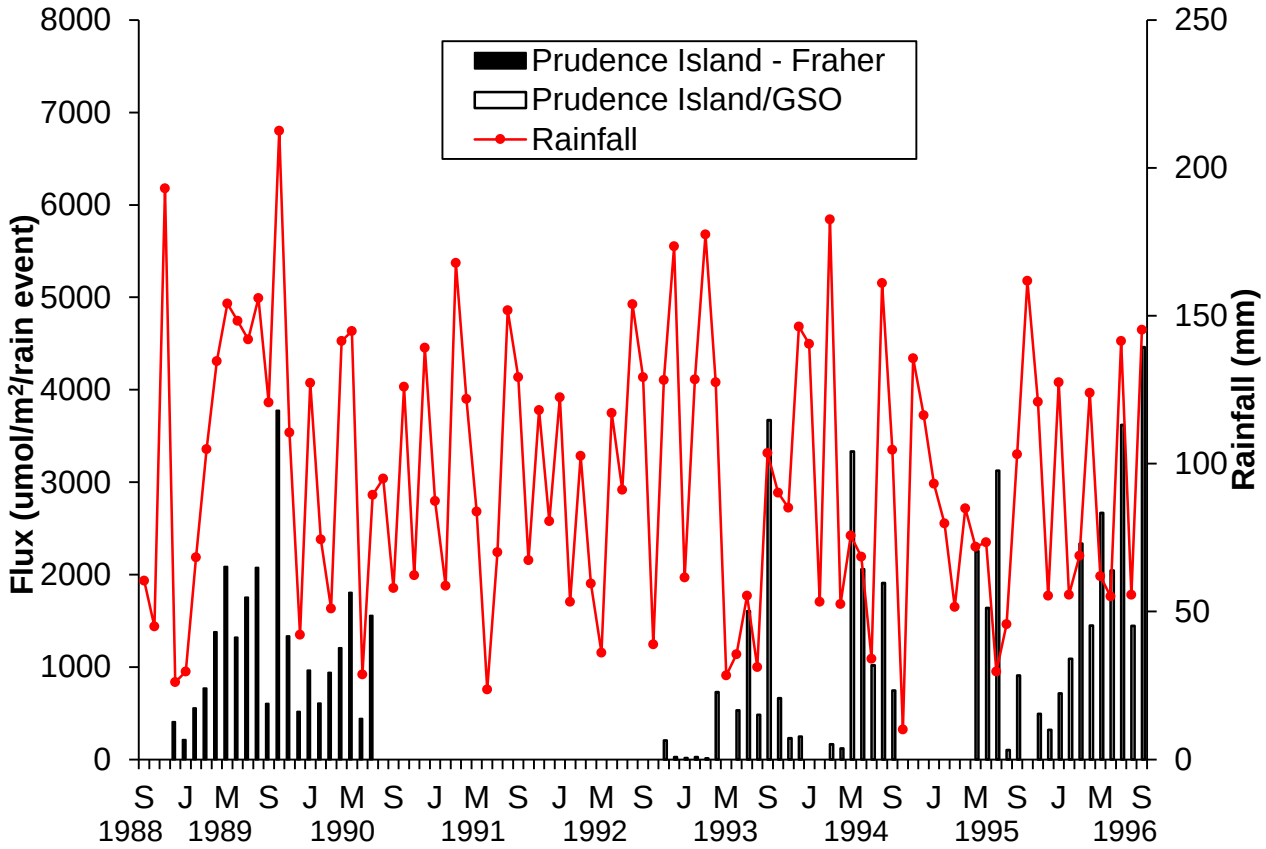

Figure 4. Flux of NH_4^+ versus month (S = September, J = January, and M = May) and year. Vertical bars represent data from 1988-1990 (left; Fraher 1991), and data from 1992-1996 (right), while the red line represents monthly rainfall totals from 1988-1996.

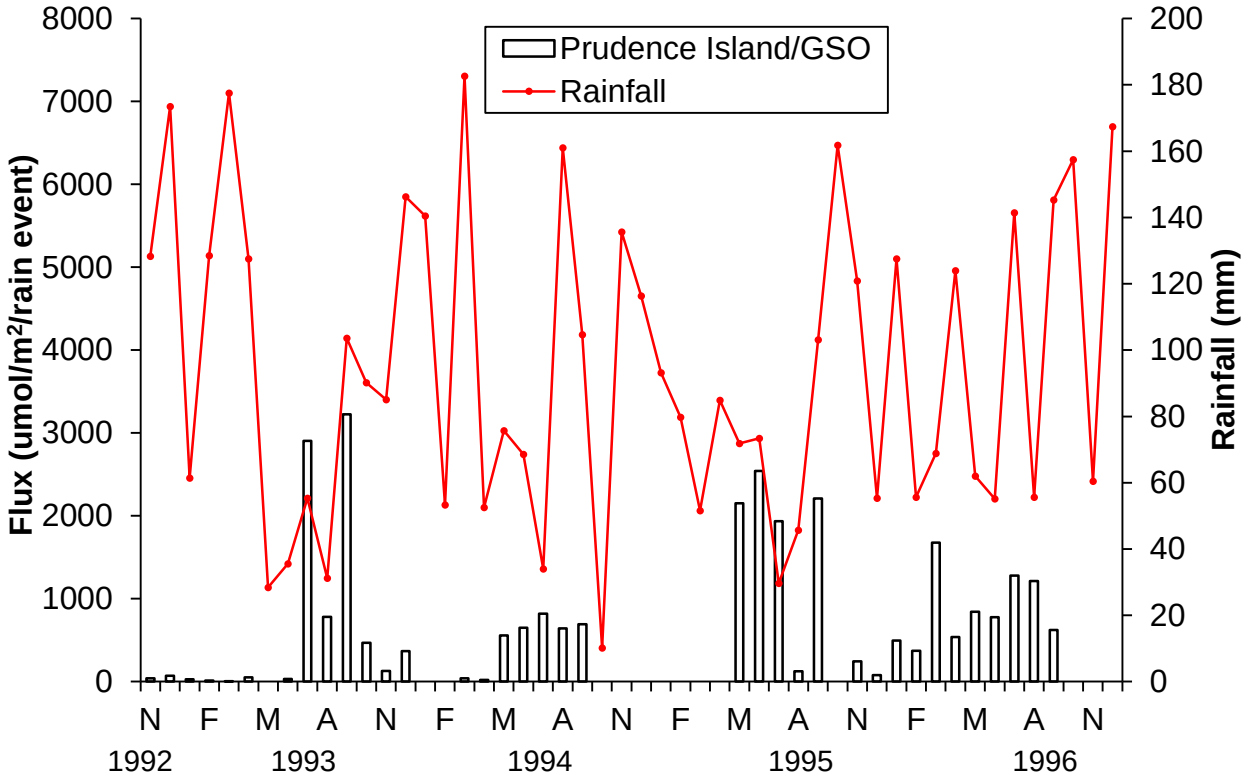

Figure 5. Flux of DON versus month (N = November, F = February, M = May; A = August) and year. Vertical bars represent data from 1992-1996, while the red line represents monthly rainfall totals from 1992-1996. No other local dataset exists for comparison.

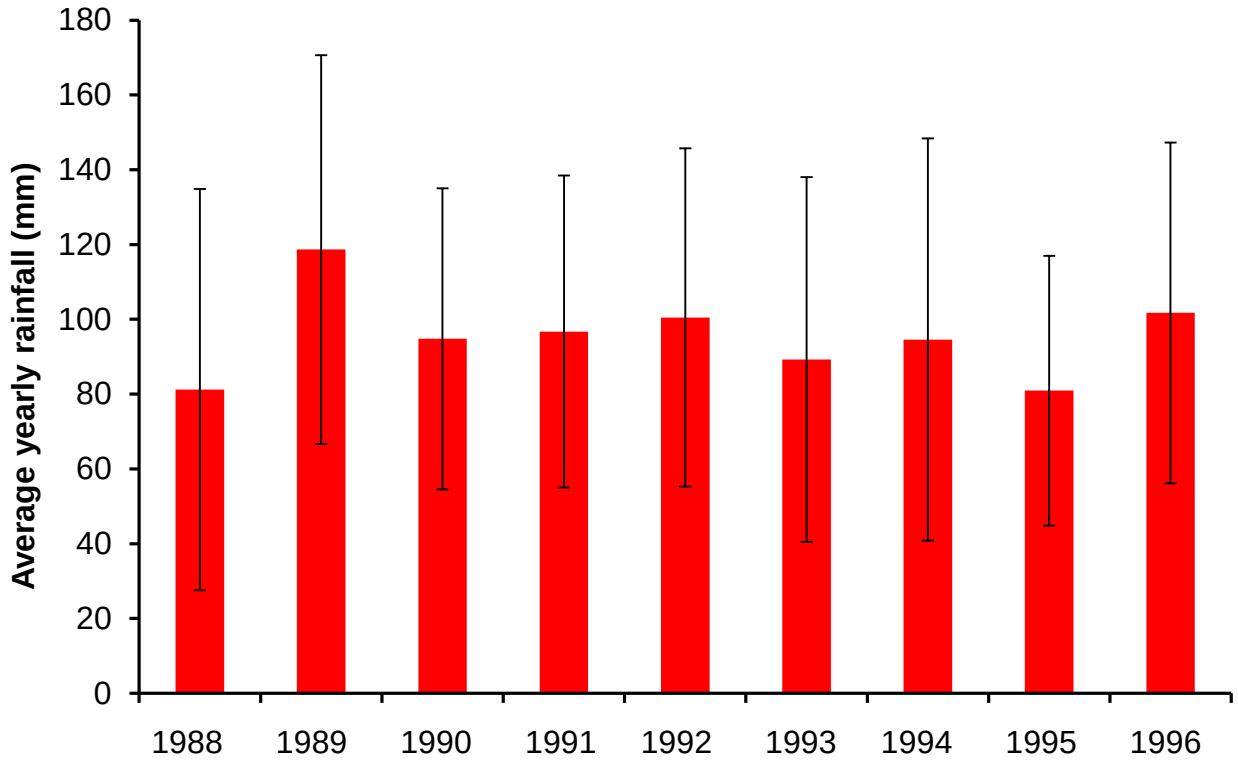

Figure 6. Average yearly rainfall with error bars.

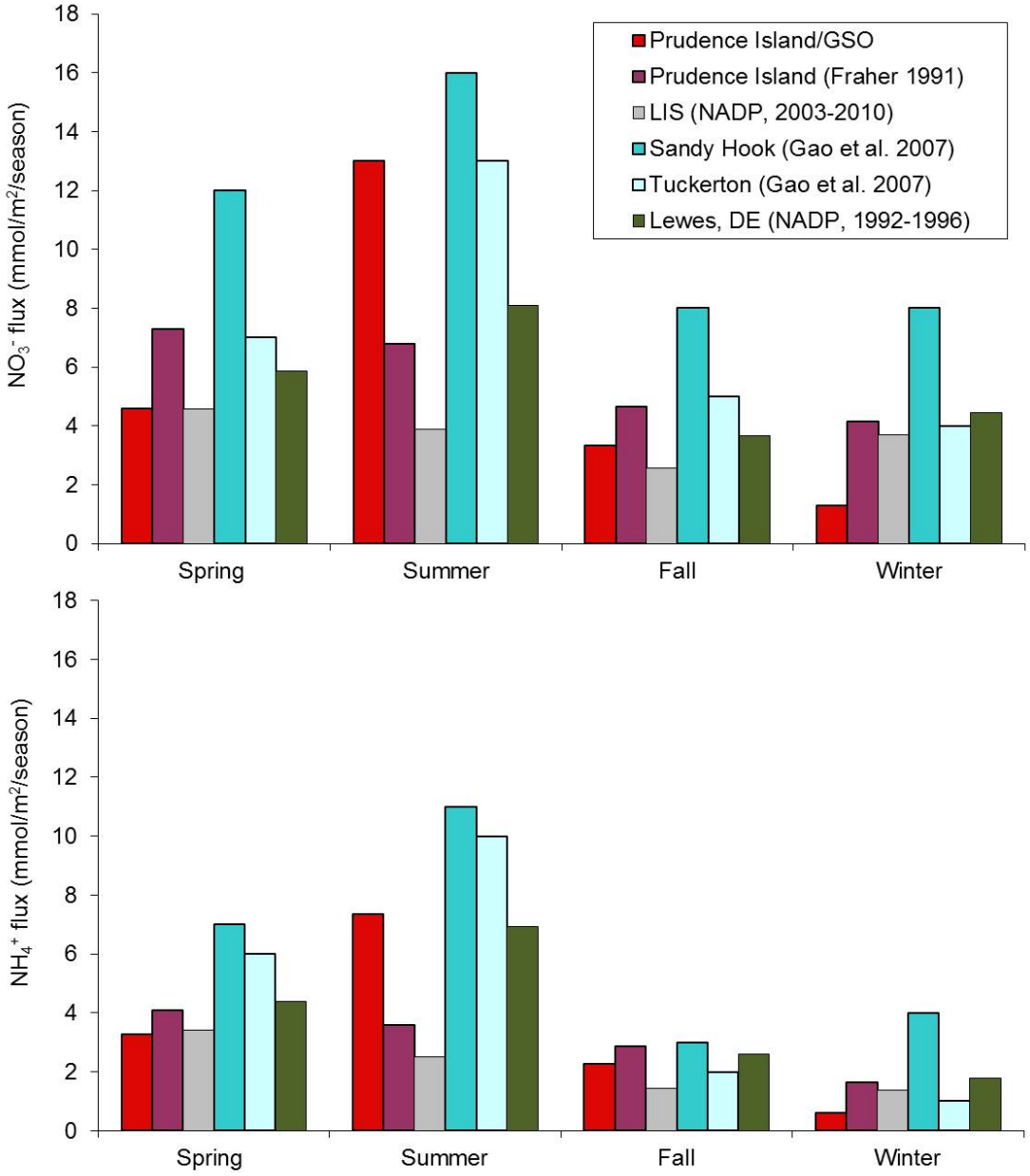

Figure 7. Regional comparison of NO_3^- and NH_4^+ flux (in mmol/m^2/season). Comparable datasets and references are listed in the legend.

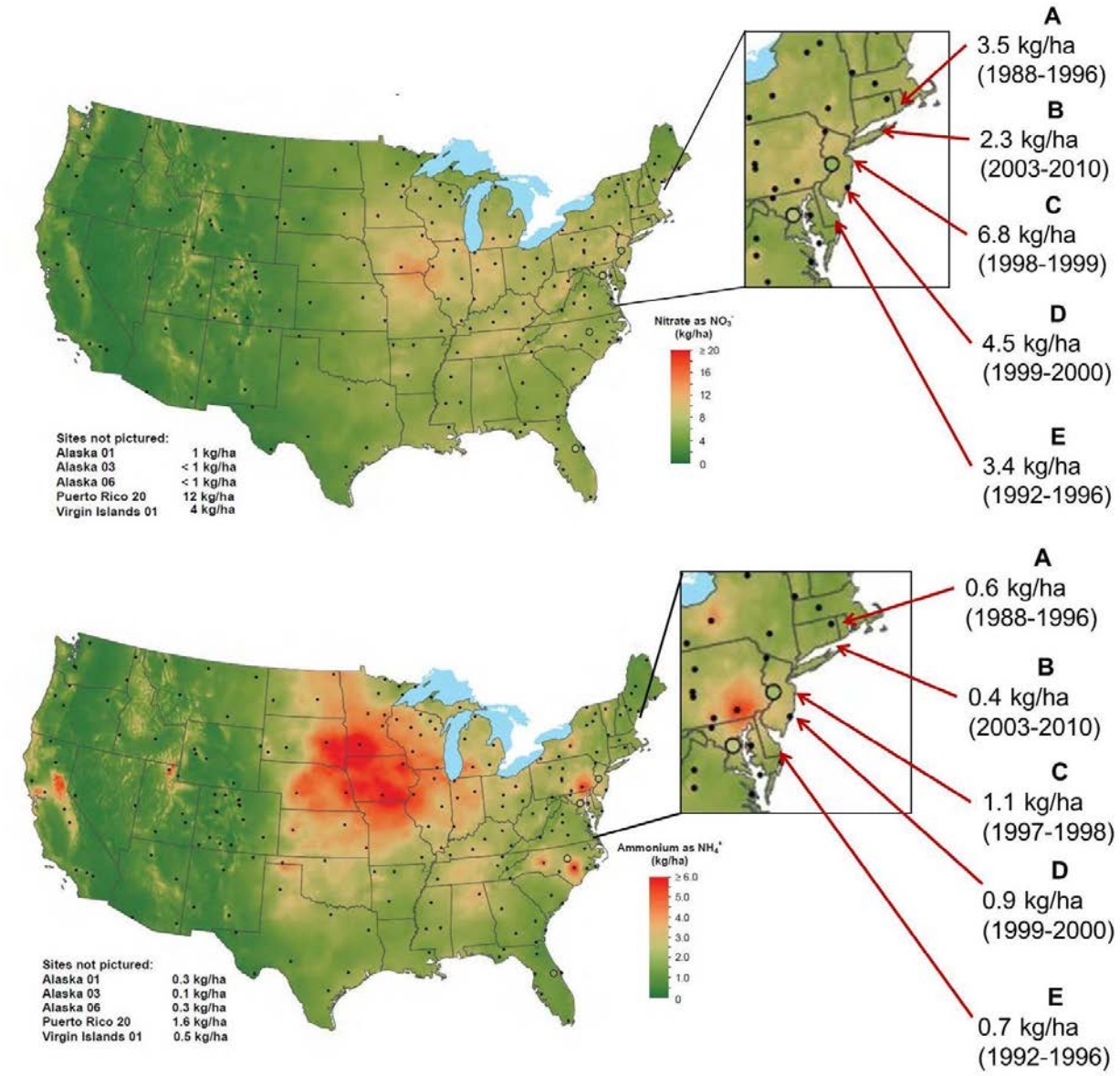

Figure 8. NO3- (top panel) and NH4+ (bottom panel) flux (in kg/ha) compared with national trends (NADP, 2010 Annual Report). A is Narragansett Bay data from this study (1992-1996) and Fraher 1991; 1988-1990); B is Long Island Sound (NADP 2003-2010); C is Sandy Hook, NJ (Gao et al. 2007; 1998-2001); D is Tuckerton, NJ (Gao et al. 2007; 1998-2001); and E is Lewes, DE (NADP; 1992-1996). All NADP data was obtained through the NADP website: http://nadp.sws.uiuc.edu/NTN/ntnData.aspx.