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Importance of atmospherically deposited nitrogen to the annual nitrogen budget of the Neuse River estuary, North Carolina

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Abstract

Wet deposition of nitrogen, as NH_4^+, NO_3^-, and organic N, contributes up to 50% of the total externally supplied or 'new' N flux to the Neuse River Estuary (North Carolina). Excessive nitrogen (N) loading to N-sensitive waters such as the Neuse River Estuary has been linked to changes in microbial and algal community composition and function (harmful algal blooms), hypoxia/anoxia, and fish kills. In a 4-year study from July 1996 to July 2000, the weekly wet deposition of NH_4^+, NO_3^-, and dissolved organic N was calculated, based on concentration and precipitation measurements, at 11 sites on a northwest–southeast transect in the watershed. Data from this period indicate that the annual mean total wet atmospherically deposited (AD)-N flux was 11 kg ha^{-1} $year^{-1}$. Deposition was fairly evenly distributed between nitrate, ammonium, and organics (32%, 32%, and 36%, respectively). Seasonally, the summer (June–August) months contained the highest weekly wet total N deposition; this trend was not driven by precipitation amount. Estimates of watershed N retention and in-stream riverine processing revealed that the AD-N flux contributed an estimated 20% (range of 15–51%) of the total 'new' N flux to the estuary, with direct deposition of N to the estuary surface accounting for 6% of the total 'new' N flux. This study did not measure the dry depositional flux, which may double the contribution of AD-N to the estuary. The AD-N is an important source of 'new' N to the Neuse River Estuary as well as other estuarine and coastal ecosystems downwind of major emission sources. As such, AD-N should be included in effective nutrient mitigation and management efforts for these N-sensitive waters.

Keywords: Atmospheric deposition; Eutrophication; Ammonia; Estuary; Nitrogen

1. Introduction

The Neuse River Estuary (NRE), part of the U.S.'s second largest estuarine complex, the Albermarle–Pamlico Sound System (Fig. 1), is an economically and ecologically valuable resource as a fin- and shellfish nursery (Copeland and Gray, 1991; Lenihan and Peterson, 1998). Over the past 20 years, symptoms of eutrophication, including nuisance cyanobacterial and dinoflagellate blooms, associated bottom water hypoxia/anoxia, fish kills, and altered food web structure have plagued this resource-rich and economically valuable system (Tedder et al., 1980; Paerl, 1983, 1987; Christian et al., 1986). The Neuse River drains a watershed of over 16,000 km^2. Forest (57%) and agriculture (26%) dominate the watershed, and urbanization is increasing. The watershed is primarily in the Coastal Plain geologic province, with the upper watershed residing in the Piedmont. Average annual rainfall in the watershed ranges from approximately 100 cm $year^{-1}$ in the Piedmont to 132 cm $year^{-1}$ in the Coastal Plain (U.S. Geological Survey, 2000). The total N load to the estuary has increased by at least 30% over the past three decades (Stanley, 1988; Dodd et al., 1993; Paerl et al., 1998), although the temporal pattern of N loadings is unclear in the past decade (Stanley et al., 1999). A complete understanding of magnitude and variability (spatial and temporal) of all N inputs is necessary to effectively manage this system from a water quality perspective.

Atmospherically deposited N (AD-N) can reach N-sensitive waterways via direct deposition to the water's surface or by deposition to land surface and subsequent runoff (indirect deposition). A portion of the indirect AD-N will be taken up by the land and will not reach the waterways. The degree to which N is retained by a land parcel depends on soil type, slope, land use, degree of fertilization, and

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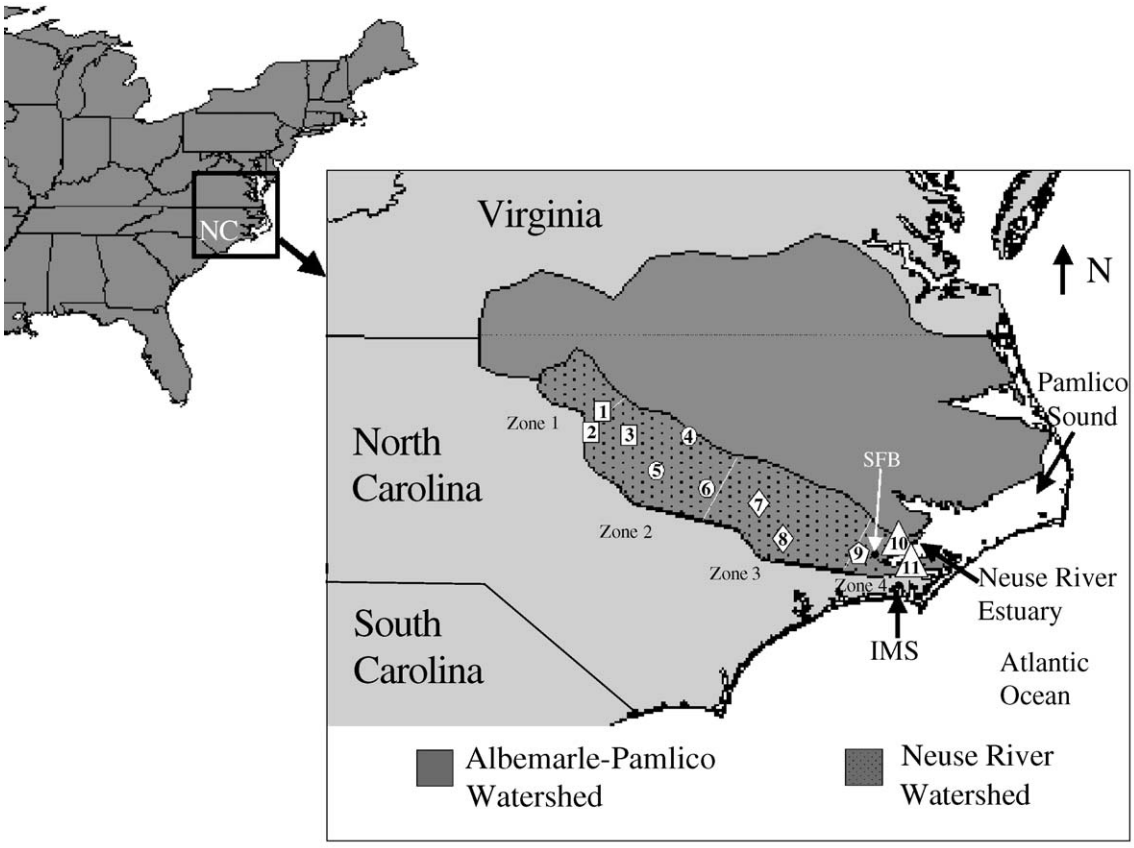

Fig. 1. Site location map. Numbers show the locations of wet atmospheric deposition of nitrogen (AD-N) collectors and rain gauges. Sites 1, 3, 4, 5, 8, and 10 are NCSCO rain gauge only sites. Sites 2, 6, 7, 9, and 11 are AD-N collector sites with rain gauges. Wet AD-N and rain gauge pairings are shown with like shapes. See Table 1 for more information. Zones shown are delineations for in-stream N degradation model (see Table 3). SFB is Streets Ferry Bridge (see text) and IMS is the UNC-Chapel Hill Institute of Marine Sciences in Morehead City.

season. Furthermore, there are two depositional pathways: wet deposition (precipitation) and dry deposition (particulate matter and gases). Due to the large uncertainties associated with measuring dry deposition, this paper will only address wet deposition.

Precipitation (rain, snow, sleet, hail) contains a variety of organic and inorganic species, including many N compounds. Many of these species are biologically available and include inorganics (NO_3^-, NO_2^-, NH_4^+) and organics (amino acids, peroxyacetlynitrate [PAN], urea) (Timpereley et al., 1985; Mopper and Zika, 1987; Duce et al., 1991; Paerl, 1995; Peierls and Paerl, 1997; Church, 1999).

In an estuarine system, 'new' N is externally supplied N-transported to the estuary in rivers or directly from the atmosphere. This includes natural sources of N such as N_2 fixation, point sources of N (municipal and industrial discharges), agricultural and urban runoff, and other nonpoint sources of N (septic fields, atmospherically deposited nitrogen that falls on the watershed [indirect deposition], and that falls directly on the estuary [direct deposition]). Total wet-deposited N flux has been shown to be an important source of 'new' N, contributing from 20% to >40% of the 'new' N flux to U.S. East Coast estuarine and coastal waters. This implicates wet-deposited N in accelerated coastal productivity and eutrophication in N-sensitive coastal ecosystems (Paerl, 1985, 1995; Asman et al., 1993).

Fossil fuel combustion is a large source of nitrogen oxides (NO_x) (Likens et al., 1974; Levy and Maxim, 1987; Duce et al., 1991) and a smaller source of NH_3 gas. While some of this NO_x may originate from local sources in eastern North Carolina, NO_x can be transported over large distances (hundreds to thousands of kilometers) due its relatively long atmospheric lifetime (1–15 days) (Aneja et al., 1998).

Sources of atmospheric organic nitrogen are not well understood. Natural sources may include sea spray droplets and pollen from plants (Prospero et al., 1996).

While NO_x is usually the most common form of dissolved inorganic nitrogen (DIN) in rainfall, in areas with intensive agriculture, NH_3 and NH_4^+ can be major DIN components as well (Buijsman et al., 1987). Ammonium is a major component of wet atmospheric N flux in eastern NC and its increase has been documented over the past 20 years (National Atmospheric Deposition Program [NADP] data; Paerl and Whitall, 1999).

Anthropogenic sources of NH_3 include stack emissions, sewage treatment plants, septic systems, and agricultural emissions (both from chemical fertilizers and animal waste).

Unlike NO_x, which can be transported over great distances, NH_3 transport tends to be on a scale of 100 km or less (Asman and Van Jaarsveld, 1992). A likely source of new NH_3–N is volatilization from animal waste. This source has increased, since 1975, with a fourfold increase in swine production and a significant increase in poultry production in eastern North Carolina (North Carolina Department of Agriculture, 2000).

The purpose of this study was to evaluate the relative importance of AD-N to the 'new' N budget for the NRE and to characterize the spatio-temporal variability of this flux.

2. Materials and methods

2.1. Sample collection, storage, and analysis

Wet deposition was sampled using a wet/dry collector (Aerochem Metrics, model 301 with co-located rain gauge) on a weekly basis (NADP sampling protocol) in a sampling transect in the Neuse River watershed (Fig. 1). Previous studies (Vet et al., 1989; Lamb and Comrie, 1993; Butler and Likens, 1998; National Oceanic and Atmospheric Administration, 1998) have reported significantly lower NH_4^+ concentrations in co-located sampling stations, when sampled weekly versus those sampled on a daily basis. This loss has been attributed to biological utilization of the N in the bucket (Sisterson et al., 1985). To address this problem, thymol ($C_{10}H_{14}O$) was used as a biocide in the wet bucket (Gillett and Ayers, 1991) to prevent biological alteration/utilization of the N species in the wet collection bucket. The collectors have covered 'wet' buckets that, in combination with the inherently low rainwater pH (4–5), reduced the potential for NH_3 loss through volatilization. All samples were stored frozen (-20 °C) until analysis via standard flow analysis techniques (NH_4^+ after Diamond and Huberty, 1996; NO_3^- from Schetig, 1997; total Kjeldahl nitrogen (TKN) from Wendt, 1997; organic N was determined by difference between TKN and NH_4^+). The use of thymol as a biocide was compatible with these techniques, but it may not be compatible with all analytical techniques and should be used with caution.

Table 1
Atmospheric nitrogen deposition sampling sites, NCSCO precipitation gauges, and pairings for deposition calculations

No.	Name	Description	Elevation (m)	DIN conc. pairing	DON conc. pairing
1	NCSU	NCSCO	121	Wake	Goldsboro
2	Wake	NADP Site	122	Wake	Goldsboro
3	Clayton	NCSCO	101	Wake	Goldsboro
4	Wilson	NCSCO	34	Goldsboro	Goldsboro
5	Smithfield	NCSCO	46	Goldsboro	Goldsboro
6	Goldsboro	UNC-IMS	33	Goldsboro	Goldsboro
7	Kinston	UNC-IMS	27	Kinston	Kinston
8	Trenton	NCSCO	9	Kinston	Kinston
9	New Bern	UNC-IMS	5	New Bern	New Bern
10	Bayboro	NCSCO	3	Beaufort	IMS
11	Beaufort	NADP/CASTNet	2	Beaufort	IMS

IMS is the UNC-Chapel Hill Institute of Marine Sciences, where wet deposition measurements were also made. These data are used for DON pairings for Bayboro and Beaufort, since NADP does not measure organic N in precipitation. See also Fig. 1.

Table 2
Nitrogen retention values for various land-use types from the literature

Model	N retention values as percent of N retained by land				
	Forest	Crop	Pasture	Urban	Other
Highest	100	99.97	99.96	95.3	75
Lowest	90	60	70	25	50
'Best' estimate	99	96	97	70	75

Values shown are highest reported, lowest reported and our 'best estimate' model. See discussion in text for derivation of 'best estimate' values.

To calculate watershed level indirect wet deposition inputs, the watershed was divided into polygons based on rain gauge locations using the Theissan Method (Schwab et al., 1993; Whitall, 2000). An array (6) of precipitation gauges, managed by the North Carolina State Climate Office (NCSCO), was used to complement the five locations in the basin that had atmospheric deposition collectors and co-located rain gauges (Fig. 1, Table 1). 'Stand alone' gauges from NCSCO were paired with concentration data from the closest wet/dry collector for deposition calculations. Two of the sites are national monitoring program sites (site 2 is a NADP site and site 11 was a Clean Air Status and Trends Network (CASTNet) site that is now part of the NADP network). Since these monitoring programs do not measure organic N, data from the closest site measuring organics were used for organic AD-N calculations. This scheme allows for the maximum resolution of spatially weighted indirect wet N fluxes based on area (the area of polygon A [km^2] × deposition flux polygon A [mg m^{-2} $year^{-1}$] = total deposition for polygon A [mg $year^{-1}$]). For direct deposition calculations, the estuary surface area was similarly divided using the Theissan method of the three wet sites that bound the estuary (sites 9, 10, and 11—Fig. 1, Table 1).

2.2. Nitrogen retention and in-stream degradation models

In order to determine the relative importance of wet-deposited N fluxes to the N budget of the estuary, we have estimated the amount of indirect deposition that would be retained by the landscape with the use of a N retention model. A problem with N retention models of this type, which use average retention values for generalized land use types, is that true nutrient retention depends on a variety of land parcel-specific parameters. These parameters include soil N content, historic acid deposition, soil type, land slope, elevation, and vegetative type (Valigura et al., 1996). By using average values for an entire watershed, amounts of the N retained in the land parcel can be calculated, but these values are not absolute and must be considered estimates.

Table 3
In stream degradation model developed by NC Department of Environment and Natural Resources Division of Water Quality

Region	% N reaching estuary
Zone 1	10
Zone 2	50
Zone 3	70
Zone 4	100

Zones are delineated in Fig. 1.

This N retention model was designed after compiling literature watershed retention values (Valigura et al., 1996; Tyler, 1988; Hinga et al., 1991; Fisher and Oppenheimer, 1991) and uses 1996 GIS land use data for the 267 subbasins in the watershed. For each land use type, a N retention value from 0 to 1 was assigned. A value of 0.25 means that 25% of the flux would be retained by the land and 75% would reach the waterways. Table 2 shows the three sets of input parameters for the model, representing the highest and lowest values reported in the literature for studies of the Chesapeake Bay. They also represent our 'best estimate' and take into account watershed specific variables such as slope, soil type, tillage, fertilizer application rate, extent of riparian buffers, and crop types for the Neuse River basin. A more complete discussion of the selection of the retention values for the 'best estimate' model is presented in Appendix A.

After the amount of wet AD-N reaching the streams from each subbasin was determined, these N loadings were applied to a simple in-stream degradation model (North Carolina Department of Environment and Natural Resources, 1993) (Table 3) to determine how much of this N reached the head of the estuary. This model accounts for N losses in the streams and rivers due to denitrification and settling as particles; N losses are proportional to distance traveled to the estuary.

3. Results

Results presented are from a 48-month period starting in July 1996.

3.1. Speciation of wet AD-N

On an annual basis, wet AD-N is fairly evenly divided between the chemical species, with NH_4^+, NO_3^-, and organics making up 32%, 32%, and 36%, respectively. The wet deposition of organic N is a potentially important component of wet atmospheric flux in coastal North Carolina (Peierls and Paerl, 1997); data from our current study support this assessment. Although the organic N fluxes reported here are higher than reported for coastal North Carolina in Peierls and Paerl (1997), they fall within published reports from other coastal areas (Timpereley et al., 1985; Knapp et al., 1986).

3.2. Variations in weekly wet AD-N flux

Seasonally, the mean total weekly wet N deposition is highest in the spring (March–May) and summer (June–August) (significant at $\alpha = 0.01$ one-way ANOVA with post-hoc Bonferroni means comparison, Fig. 2). This pattern is mirrored in the seasonal patterns for both NH_4^+ and NO_3^- deposition (data not shown). However, this pattern does not persist in seasonal precipitation patterns; seasonally, spring has the lowest average weekly precipitation (Fig. 2). This suggests that other factors, including the direction from which a storm system originates and/or passes over N source regions and seasonal changes in sources, may be involved in causing these differences.

3.3. Spatial variability in annual wet AD-N flux

On an annual basis, the highest wet AD-N flux occurs in the middle segment of the watershed, with the lowest fluxes occurring in the upper watershed. This pattern is driven by spatial differences in NO_3^- deposition (Fig. 3); deposition of NH_4^+ and organics do not vary spatially (data not shown). Rainfall amount does not significantly vary across the watershed, although on an annual basis, coastal sites receive slightly more rain than inland sites.

3.4. Total annual deposition

Total N deposited from the atmosphere, due to wet deposition to the land area of the Neuse River Basin, was estimated to be 16.9 Gg N/year (standard deviation ± 6.6 Gg) or 11 kg ha^{-1}. Using a nitrogen retention model and in-stream riverine degradation model (see discussion above and Tables 1 and 2), we estimated the amount of wet AD-N

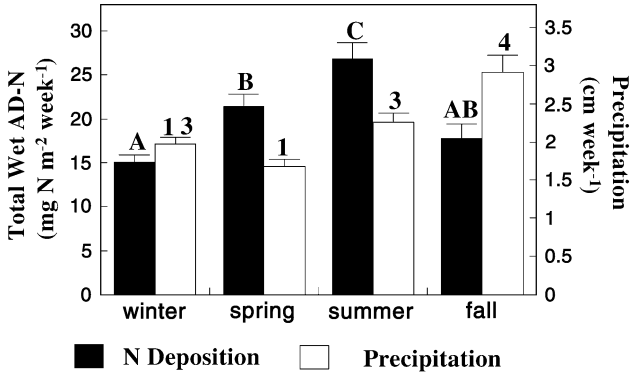

Fig. 2. Seasonal variability in weekly wet nitrogen deposition (black bars) and precipitation depth (white bars) for 11 sites pooled. Letters show significant differences between deposition groups and numbers show significant differences between precipitation groups (one-way ANOVA with post-hoc Bonferroni analysis, $\alpha = 0.05$). Groups with common letters or numbers are not significantly different from each other.

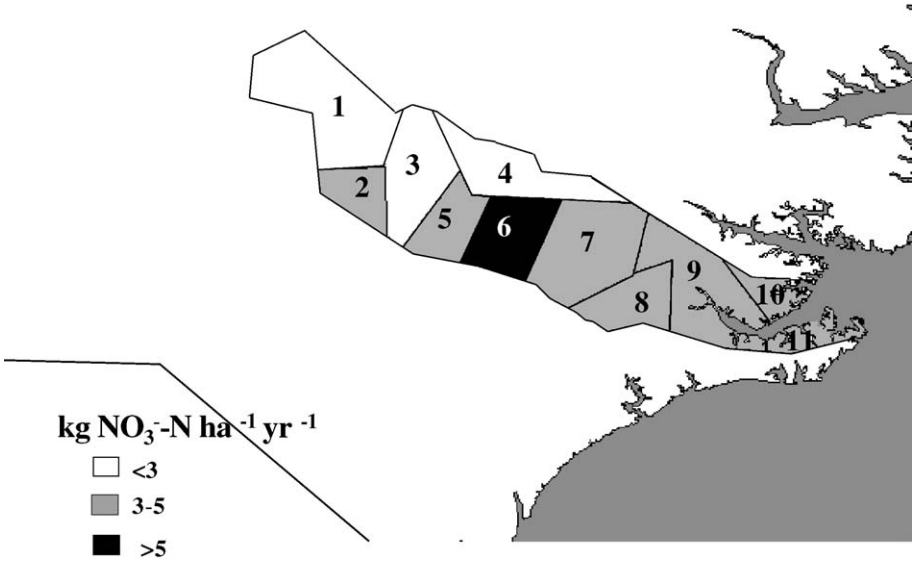

Fig. 3. Spatial variability in atmospherically deposited nitrate. Data shown are 4-year average deposition (July 1996 to July 2000). Deposition for polygon 6 is statistically greater (one-way ANOVA with post-hoc Bonferroni analysis, $\alpha = 0.05$) than polygons 1, 2, 3, and 4.

retained in a land parcel, based on land use type, and how much of the N reaching the waterways was transported to the estuary. This flux ranged from 0.7 to 3.7 Gg N year^{-1}, depending on the nutrient retention values used (Table 4). Using our 'best fit' values, this flux via indirect deposition was estimated to be 1.0 Gg N year^{-1}.

Direct deposition to the estuary surface (based on sites 9, 10, and 11 that bound the estuary) was 0.4 Gg N year^{-1} (standard deviation ± 0.8 Gg), resulting in a total wet AD-N flux to the estuary of between 1.1 and 4.1 Gg N year^{-1} ('best' estimate = 1.5 Gg N year^{-1}). The total flux of nitrogen to the estuary is 7.5 Gg N year^{-1} (riverine loading above New Bern, NC at Streets Ferry Bridge [SFB], Lebo, personal communication; Fig. 1). Thus, the atmospheric fluxes represent between 15% and 55% ('best' estimate = 20%) of the total 'new' nitrogen flux to the estuary (Table 4). The total flux inherently includes indirect wet AD-N, directly deposited wet AD-N, point source loading (N.C. Department of Environment and Natural Resources, personal communication, 2000), and nonpoint source loading (NC Department of Environment and Natural Resources, 1993) below the SFB.

Table 4
The relative contribution of AD-N to the 'new' nitrogen budget of the Neuse River Estuary

Model	Direct AD-N flux to estuary (Gg year^{-1})	Indirect flux reaching estuary (Gg year^{-1})	Total AD-N to estuary (Gg year^{-1})	AD-N flux as percent of total 'new' N flux to estuary (%)
Highest	0.4	0.7	1.1	15
Lowest	0.4	3.7	4.1	55
Best 'Estimate'	0.4	1.0	1.5	20

4. Discussion

The data presented here indicate that wet-deposited N is fairly evenly distributed between the chemical species. From an ecological response perspective, however, the inorganic species may be more important on short time scales than the organic species due to their high degree of availability to phytoplankton (Antia et al., 1991; Peierls and Paerl, 1997). Previous studies (Paerl and Whitall, 1999) using National Atmospheric Deposition Program (NADP) data from eastern North Carolina (site NC35) have indicated that NH_4^+ has contributed to an increasingly larger percentage of the inorganic wet AD-N budget over the past two decades. The relatively short duration of this study precludes us from making similar conclusions.

The seasonal variability in weekly wet AD-N flux cannot be explained by variability in precipitation amount alone (Fig. 2). Therefore, this seasonal pattern must be explained by other variables such as seasonal fluctuations in source emissions of atmospheric N and other meteorological factors. For example, during the summer, elevated air temperatures lead to relatively high NH_3 volatilization rates from animal waste stored in lagoons or applied to land (Aneja et al., 2000).

This study is one of the first efforts to quantify the importance and patterns in wet AD-N on a watershed scale for a N-sensitive coastal watershed. Nitrogen in precipitation is clearly an important component of the N flux to the Neuse River Watershed, accounting for between 15% and 55% of the total 'new' nitrogen flux to the estuary. The direct depositional component that makes up 4–6% of the total 'new' N loading may be particularly important, when considering wet AD-N's contribution to the 'new' N budget of Pamlico Sound, due to its large surface area (4500 km^2).

This study did not address the dry deposition of nitrogen, which may be a significant additional flux of N to the system. Two of the U.S. Environmental Protection Agencies' Clean Air Status and Trends Network (CASTNet) sampling sites are located in eastern North Carolina. Modeling and monitoring efforts at these two sites estimate the dry deposition of particulate NO_3^- and NH_4^+ and gaseous HNO_3^- to be between 1.5 and 3.2 kg N ha^{-1} $year^{-1}$ (80% as oxidized N) for the time period of this study (United States Environmental Protection Agency, 2000). This would represent up to a 33% increase over the basin-wide average N deposition reported here. It should be noted that the CASTNet data do not include deposition of gaseous NH_3 that may be quite important in the Neuse River watershed due to agricultural activities.

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Appendix A

The values for the 'best estimate' model attempt to further refine the published range of values for the U.S. East Coast estuaries come primarily from studies in the Chesapeake Bay watershed. The parameters that influence N retention include soil N, groundwater characteristics, historical acid deposition, soil type, land slope, elevation, and vegetative type. Some of these parameters differ between the Neuse region and the Chesapeake Bay region.

In general, the Chesapeake Bay Watershed has steeper slopes than the Neuse River Watershed (Trapp and Horn, 1997). The steeper slopes mean a higher runoff rate and the potential for more nitrogen to be transported from land to water, when compared to areas of lower slopes (Schwab et al., 1993). Therefore, based on average slope, the Neuse River watershed should have relative high retention values compared to the Chesapeake Bay watershed.

The extent to which forests retain N is in part determined by historical AD-N loading (Gundersen et al., 1998; Tietema, 1998). Based on NADP data for five sites in or near the Neuse River watershed and six sites in or near the Chesapeake Bay watershed, the Chesapeake Bay watershed has a higher historical annual AD-N flux than the Neuse (Whitall, 2000). This means that forests in the Neuse will generally retain more N than forests in the Chesapeake.

One potential mechanism for N retention is loss to groundwater. Groundwater in the Chesapeake Bay watershed has a much longer retention time ($T_{res} = 200$ years) (Bohlke and Denver, 1995) compared to that of the Neuse River watershed ($T_{res} = 50$ years) (Reynolds and Spruill, 1995). Therefore, in general, the Chesapeake Bay watershed will have greater N retention based on groundwater retention time.

Based on these general differences between the Neuse River watershed and the Chesapeake Bay watershed, we chose N retention values that approach the maximum reported retention values for the Chesapeake (Table 3). Three sets of parameters are shown in an attempt to bound our 'best estimate' model with maximum and minimum values.

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