Appendix VI

Atmospheric Deposition Literature Review for the Neuse River Basin (March 2008)

Summary of the Current North Carolina Atmospheric Nitrogen Deposition Literature.
Nora Deamer – DENR-DWQ-Planning Section-Basinwide Planning Unit
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The Division of Water Quality has attempted to reduce coastal eutrophication in many river basins throughout NC by managing nitrogen (N) inputs to these N-Sensitive ecosystems. Among the many different sources of anthropogenic nitrogen, the Divisions management strategies attempts to reduce nitrogen loading from both point source discharges (municipal and industrial) and from many nonpoint sources such as agricultural runoff and stormwater from new development.

The Division determined that a 30% reduction in nitrogen loading at New Bern would be needed in order for the Neuse River Estuary (NRE) to meet its designated uses and no longer suffer from excessive algal blooms and chlorophyll a exceedances (less than 10% of the samples collected over a 5 year time period would have a chlorophyll a value greater than 40 μg/l).

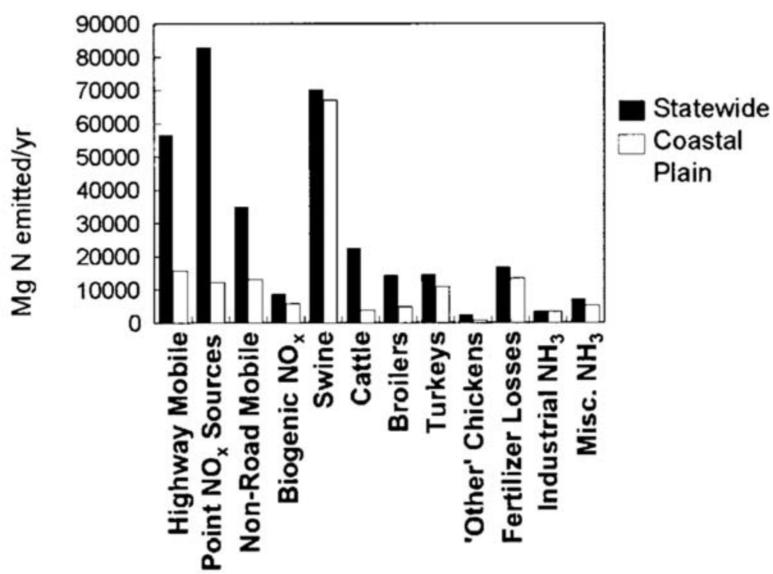
The 2006 water quality assessment of the NRE indicates it is not meeting its designated uses due to excessive chlorophyll a standard violations throughout much of the estuary. The Neuse Management Rules aimed at activities to reduce nitrogen by 30% from both point and nonpoint sources were only fully implemented in 2003 (time frame set in the strategy allowing time for the discharges to come into compliance with the rule). Based on current accounting methods, both the point and nonpoint discharges have achieved or even exceeded the 30% reduction target in nitrogen discharged to the river.

However, as of 2006, loading estimates at New Bern (Fort Barnwell – station/STORET # J7580000) are not reflecting the calculated reductions implemented by both the point and nonpoint source dischargers. There are several possible reasons why this may be occurring. One possibility is due to the excessive amount of nitrogen currently recycling in the system. It could take several years to possibly decades before the reductions are seen in the loading estimates and instream concentration reductions (1,20). It is also possible that there are sources of nitrogen that are continuing to flow into the system, as well as, sources that were not accounted for in the original loading calculations such as atmospheric deposition and groundwater (base flow) nitrogen contributions. In reviewing the current scientific literature, it appears that atmospheric nitrogen deposition is likely a highly significant and increasingly important source of "new" nitrogen to the NRE and other coastal systems along the east coast.

General information

In the past century, atmospheric nitrogen emissions/deposition has increased tenfold due to the increases from urbanization, industrial expansion and agricultural intensification (1,4). It is estimated that from 10 to greater than 40% of new nitrogen (N) supplied to these N-sensitive waters is of atmospheric origin (1,2,4). In mid-Atlantic region the atmosphere is likely a dominant source of new N (1).

There are two depositional pathways for atmospheric N deposition: wet deposition (precipitation) and dry deposition (particulate matter and gases). Nitrogen is deposited in a variety of organic (amino acids, urea, organonitriles, N-heterocyclics) and inorganic (NO_3^-, NO_2^-, NH_3, NH_4^+) forms, most of which will have a direct impact on eutrophication and the productivity of coastal waters (2,3,4,9,19). Most of the studies available to date have only assessed the contribution from wet deposition. The assessment of dry deposition is much more difficult and has only recently been established at many of the National Atmospheric Deposition Program (NADP) or Division of Air Quality (DAQ) stations throughout NC. Atmospheric deposition enters the aquatic system via direct deposition to the water surfaces or indirectly via land deposition that ultimately reach surface waters through runoff or groundwater. A portion of the indirect deposition will be utilized prior to reaching surface waters. The exact amount varies depending on the land use, soil and vegetation type, fertilization rates, slope of the land, and time of year the deposition occurs (2).



Relative contribution of various nitrogen (N) sources to the atmospheric N emissions budget for North Carolina (black) and coastal North Carolina (white). Statewide N emissions total 334 748 Mg/yr (coastal plain total = 155 931 Mg/yr). From North Carolina Department of the Environmental and Natural Resources Division of Air Quality (1996).

Figure 1 - From the *Technical Report - Atmospheric Pollutants and Trace Gases - Spatiotemporal Variability of Wet Atmospheric Nitrogen Deposition to the Neuse River Estuary, NC* (3).

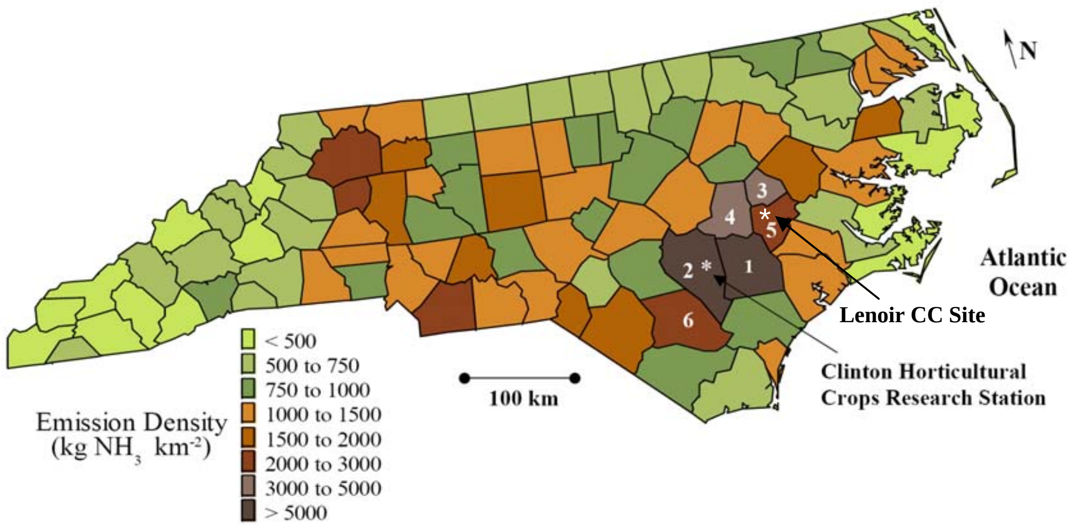
While nitrogen oxides (NO_x), generated mostly by fossil fuel combustion, accounts for the majority (50-75%) of the nitrogen pollution in the US, ammonia (NH_3) and ammonium (NH_4^+) have become the most abundant form of the atmospherically deposited nitrogen in areas with intensive agricultural operations like in the Midwest and Mid-Atlantic regions (2,4). Anthropogenic sources of NH_3 include industrial emissions, sewage treatment plants, septic systems, and agricultural emissions from both chemical fertilizers and animal waste (2,5). Figure 1 from Whitall and Paerl (2001) represent a graph of the relative contribution of various N sources to the atmospheric N emissions budget for statewide and coastal NC (data was from DAQ 1996) (3).

Since the 1980's agricultural ammonia emissions have become one of the major worldwide air pollution problems and have attracted more and more attention from the public and government regulators (4,10). Globally, domestic animals are the largest source of atmospheric NH_3 (5). A 1999 DAQ report indicated that in Europe, over 92% of ammonia emitted originated from agricultural sources (9). Of the 92%, about 80% was associated with the decomposition of livestock manure and the rest with the volatilization of fertilizer-N (9).

Walker et al. (2004) reported that livestock and fertilizers represented 78% and 10% of the total (131,500 ton NH_3-N) NC statewide NH_3 emissions respectively (5). Based on 1996 estimates for the State of NC, swine production accounted for 46.5% (77,700 tons NH_3-N) of all estimated NH_3-N emissions (167,200 tons NH_3-N) and within the 6 county area in Eastern NC with the highest swine population densities, swine production accounts for approximately 80% (57,679 tons NH_3-N) of the estimated emissions from domestic animals (see Table 1 for top 10 county hog production) (11). Figure 2, modified from Walker et al. (2004) depicts the county level NH_3 emission density for the State of North Carolina (5).

Table 1. 2006 Top 10 Counties by hog production (USDA - Agricultural Statistic)

Counties	**Production Number**	**Main River Basin**
Duplin	2,167,000	Cape Fear
Sampson	2,025,000	Cape Fear
Bladen	815,000	Cape Fear
Wayne	515,000	Neuse
Robeson	353,000	Lumber
Greene	350,000	Neuse
Lenoir	312,000	Neuse
Pitt	280,700	Tar-Pamlico
Columbus	244,000	Lumber
Pender	237,000	Cape Fear



County-scale NH_3 emission density for North Carolina, along with measurement site. Livestock activity data represent 2000 levels. All other activity data represent 1996 levels. Counties within the NH_3 source region surrounding the site are numbered according to total emissions (kg NH_3 km^{-2}): 1. Duplin, 7580; 2. Sampson, 5825; 3. Greene, 4157; 4. Wayne, 3943; 5. Lenoir, 2769; and 6. Bladen, 2002 kg NH_3 km^{-2}.

Figure 2. Modified figure 1 from *Ambient ammonia and ammonium aerosol across a region of variable ammonia emission density.* Atmospheric Environment 38 (2004) 1235-1246. JT Walker, Dave Whitall, Wayne Robarge, Hans Paerl.

In 1997, the NC DAQ examined the statewide distribution of the annual average ammonium ion (NH_4^+) concentrations from 1978 to 1997 for the NADP/NTN (National Trends Network) sites (including two collocated sites and the Great Smoky Mountain, Tennessee site). The average stayed about 0.2 mg/l until 1989. From 1990 to 1997 there appeared to be an increase of 17%. They also reported that ammonium ion concentrations in the wet deposition increased significantly at the NADP Sampson County station (NC35) over this same time period (see Figure 3) (8). This coincided with a rapid 274% increase in the statewide swine population and a 295% increase in the Sampson County population between 1989 and 1997 when this increase in deposition was reported by DAQ (USDA- National Agricultural Statistic Service; see Figure 4). In a follow up DAQ 1999 “Status Report on Emissions and Deposition of Atmospheric Nitrogen Compounds from Animal Production in NC” they estimated that the swine, poultry and cattle comprised 20.3%, 11.2%, and 7.4% respectively, of total atmospheric nitrogen emissions in NC (9).

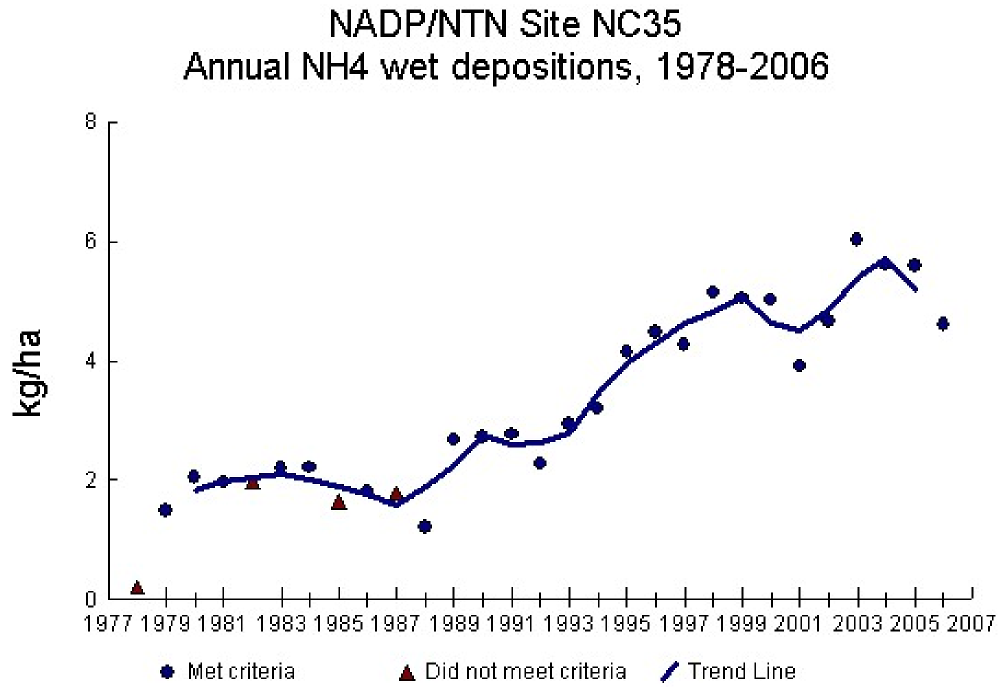


Figure 3. Annual NH_4^+ wet deposition at the Sampson County NADP station (NC35).

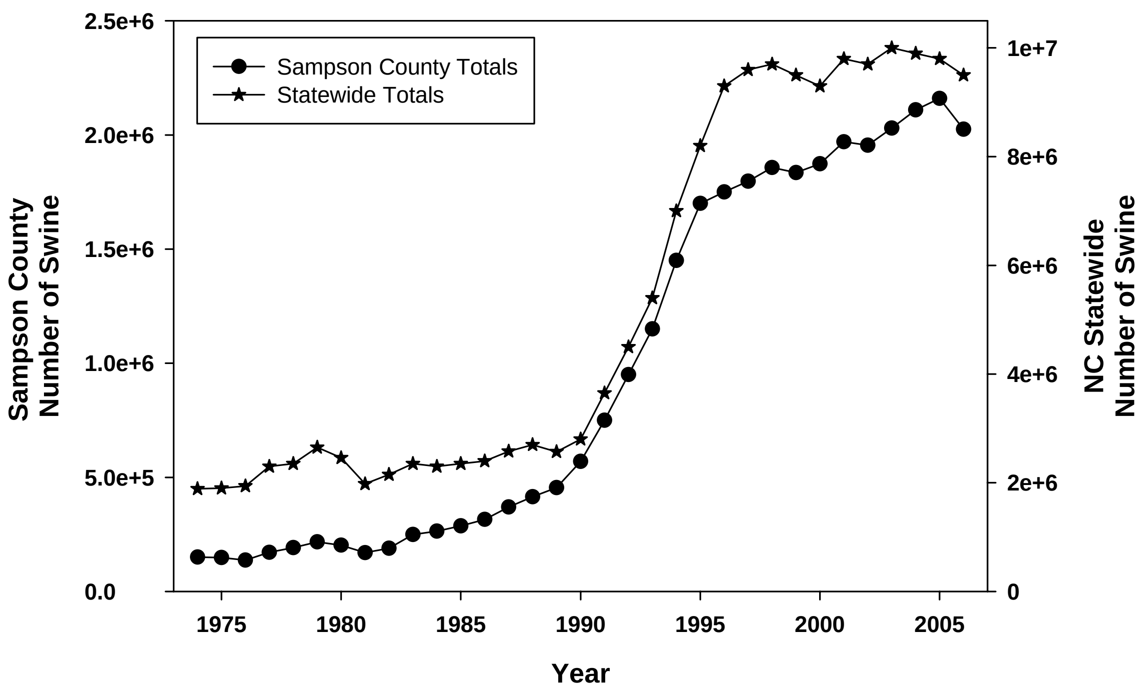


Figure 4. USDA swine statistics for the State of NC and Sampson County.

The swine industry in NC had become a very important factor in the state's economy and experienced tremendous growth until 1997 when a moratorium went into place restricting the new construction or expansion of existing farms (7). The majority of the growth in the swine industry occurred in the southeastern region of the state. Three of the top 10 hog producing counties are located in the Neuse River basin while the rest mostly lie to the south in the Cape Fear and Lumber basins (see Table 1). All of these are located within the airshed of the Neuse River Basin, resulting in a large deposition of nitrogen into the Neuse due to wind patterns out of the southwest for a large portion of the year (9). An airshed for a particular body of water can encompass hundreds of miles and is defined as the geographic area that contains the emissions source that contributes 75% of the pollutants deposited in a particular watershed (4). The main sources of ammonia emissions from these CAFOs (concentrated animal operations) are the animal barns, the waste treatment lagoons, sprayfield application and the revolatilization from the soils after spraying (9).

Atmospheric Research Station Information

The two NADP and DAQ stations that were used to determine the potential impact from CAFOs on the Neuse ecosystem were the Sampson County Clinton Crop Research Station (NC35) and the Kinston County Lenoir Community College Station (see Figure 2 for station location and Figures 4 and 5 for corresponding swine populations).

Sampson County Station

Hog farms surround the Sampson County, Clinton Crop Research Station (NC35). Changes in the annual NH_4^+ and NO_3 wet deposition and annual precipitation, at the Sampson County Station between 1978-2006 can be seen in Figures 3, 6, and 7 (18).

Depositional changes are reflected in the long-term dataset, with an increase in wet NH_4^+ deposition, increasing from about 2 kg/ha in the early 1980's to between 4 and 6 kg/ha in the mid 2000's. The deposition of wet NO_3 has varied slightly over this same time period. Paerl et al. (2002) also reported NH_4^+ deposition has increased relative to NO_3^- deposition, reflecting a national trend in agricultural regions. The increasing ratio reflects the rapid growth in intensive animal operations and simultaneous controls of NO_x emissions from burning of fossil fuels (1).

Wayne Cornelius (1997) from DAQ compared nitrogenous ion wet deposition and animal census trends to determine whether concentrations of nitrogen ions vary in association with surrounding area population densities. He found that in Sampson County there is an upward trend in the ammonium concentration that appears to begin in 1988 or 1989, along with the upward trend in hog population beginning as early as 1982, and becomes very steep from 1989 onward (Figures 8 and 4) (7). The annual mean NH_4^+ concentration exhibited a very high significant positive rank correlation with Sampson County hog densities ($r = 0.81$, $p = 0.0008$) (7). He also noted a positive correlation was also found between NH_4^+ concentrations and hogs when hog densities were greater than

140 hogs/sq mile (7). Several others also reported statistically significant increases in the wet ammonium-N concentration within this highly intensive swine producing region of NC (11,12).

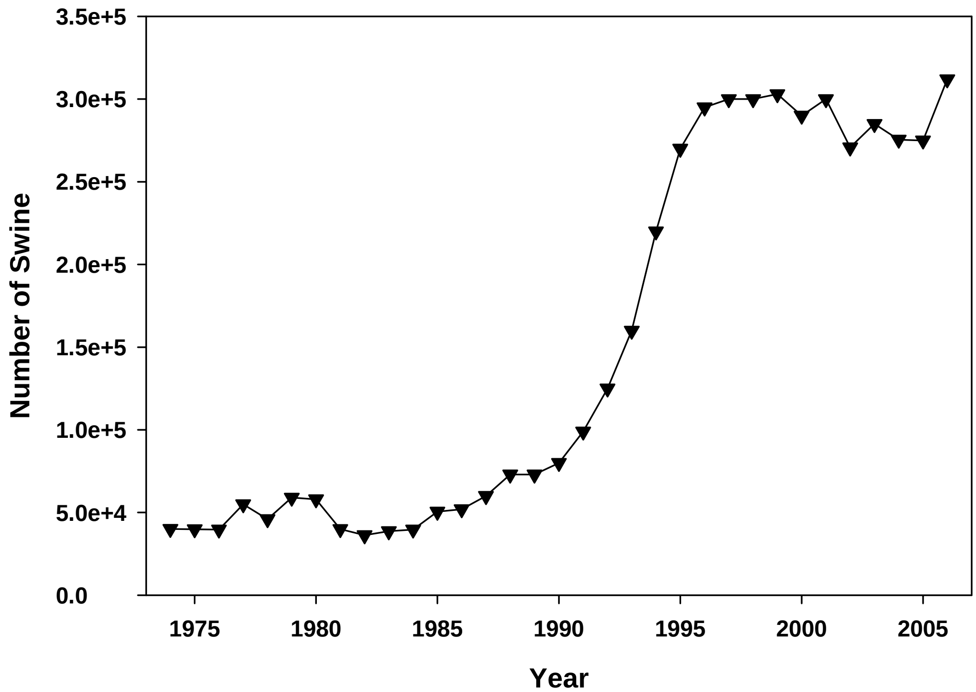


Figure 5. USDA swine statistics for Lenoir County (note the different scale used from Figure 4).

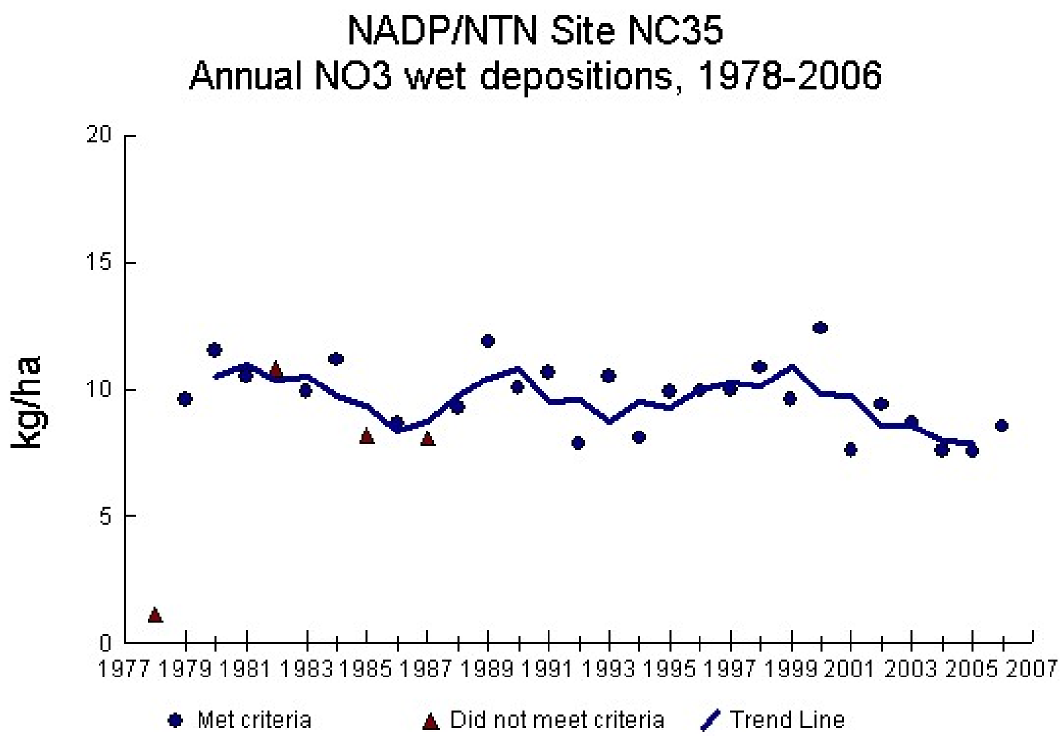


Figure 6. Annual $NO3^-$ wet deposition at the Sampson County NADP station (NC35).

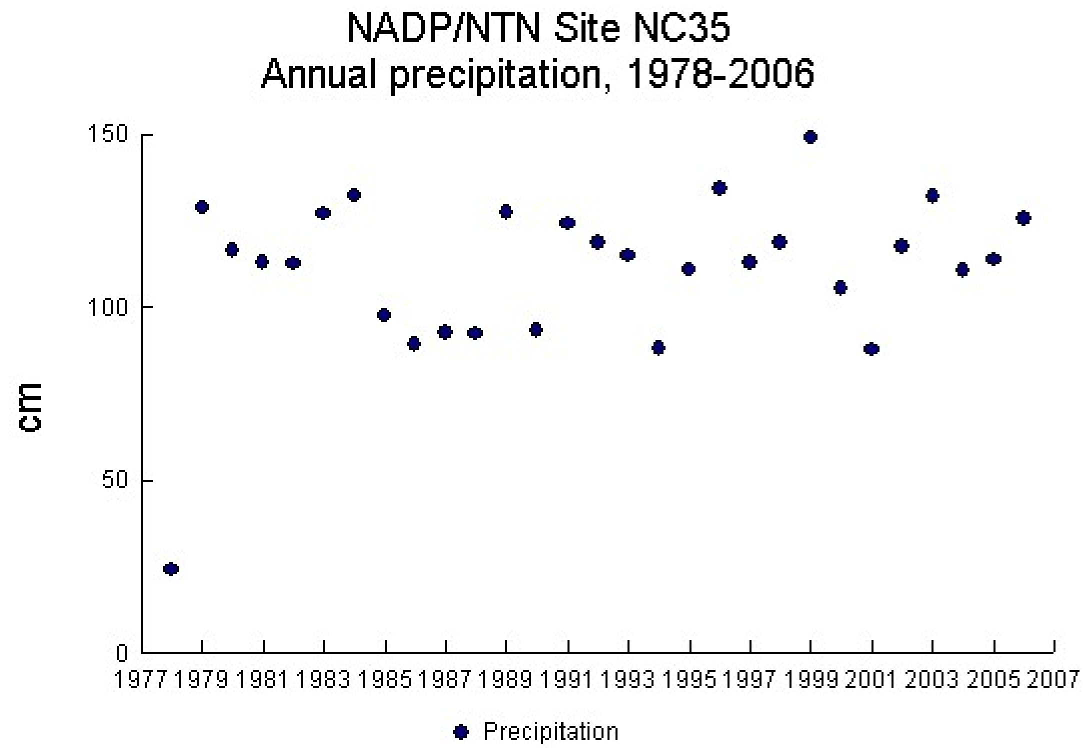


Figure 7. Annual precipitation at the Sampson County NADP station (NC35).

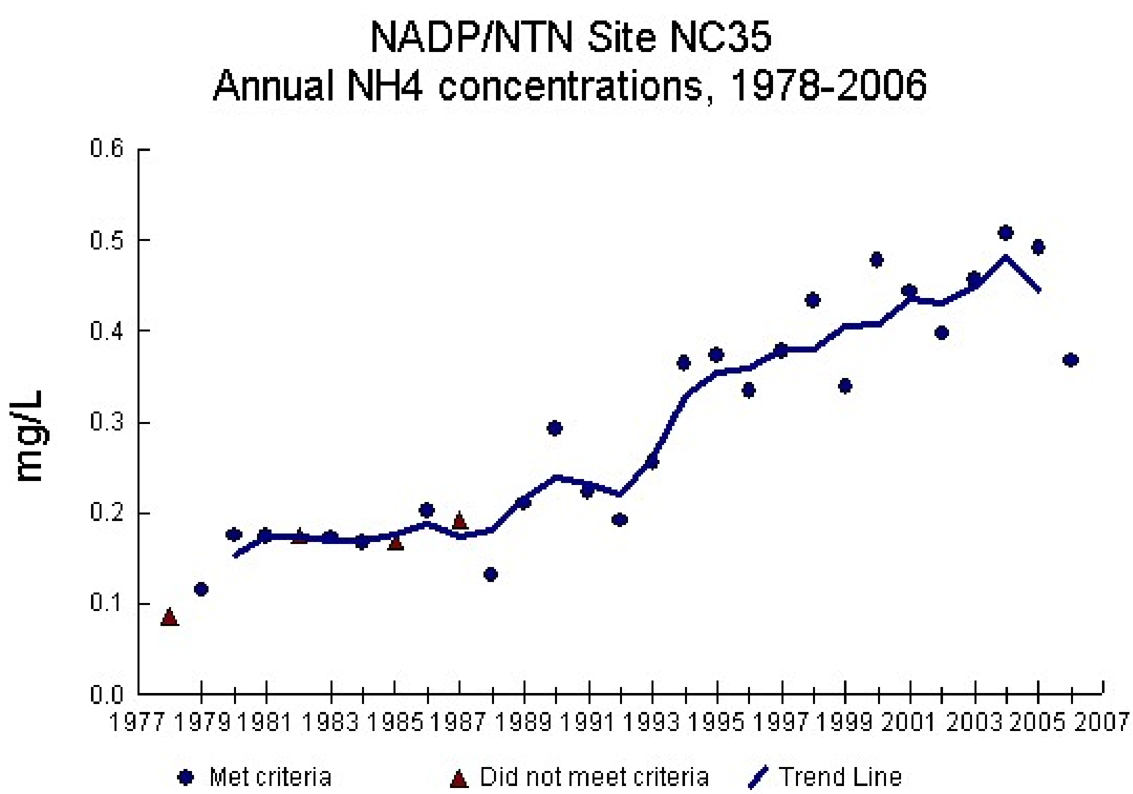


Figure 8. Annual NH_4^+ concentration at the Sampson County NADP station (NC35). .

Kinston County Station

The Kinston County, Lenoir Community College Station is approximately 50 miles northeast of the Sampson County station (10). This station is also in an area with a large hog population, however much less than the Sampson County station (Table 1 and Figure 2).

Large seasonal variations in wet atmospheric N deposition were seen at the Kinston County station (Table 2). Seasonal patterns are typical as increased summer temperatures lead to increased volatilization of NH_3 rates from animal waste storage lagoons and applied crops (5). Winds at Kinston site are primarily from the southwest during spring and summer and from the north-northeast during fall and winter (5). The higher spring and summer deposition rates may result primarily from the increased emission rates from local agricultural NH_3 sources as well as through transport from downwind sources to the southwest during the spring and summer (5). Kinston experienced higher NH_3 concentration during the late afternoon suggesting that the transport of NH_3 from downwind sources maybe the cause of the higher daytime concentrations as well. (5)

Table 2. Modified table 3 from *Ambient ammonia and ammonium aerosol across a region of variable ammonia emission density* (5).

Seasonal median NH_3 and NH_4^+ concentrations ($\mu g\ m^{-3}$), with number of observations in parenthesis, along with the ratio of summer to fall median concentrations.

		Clinton	Kinston
NH_3	Winter	2.59 (155)	0.49 (24)
	Spring	4.62 (115)	3.93 (40)
	Summer	6.18 (166)	2.72 (106)
	Fall	2.85 (130)	0.86 (96)
	Summer/Fall	2.17	3.16
NH_4^+	Winter	1.88 (155)	1.56 (24)
	Spring	1.69 (115)	1.14 (40)
	Summer	1.69 (166)	0.98 (106)
	Fall	1.45 (130)	1.23 (96)
	Summer/Fall	1.17	0.80

Dry N deposition

Unfortunately, very little is currently known about the concentrations of dry nitrogen deposition in NC. As with wet deposition, dry deposition rates are likely to vary across the state depending on the proximity to local sources. DAQ has found that accurately assessing the importance of the dry deposition of ammonia in eastern NC has so far proven to be the most problematical aspect of the their efforts to develop and evaluate regional and local scale models of ammonia emissions and deposition. There is not a regional database for the study of dry N deposition, which for a "sticky" molecule like ammonia, will vary substantially with the distance from the source and the roughness of the surrounding land cover (9).

DAQ started collecting dry N deposition data at several stations in eastern NC in the early 2000s. DAQ reported on a short term dry deposition study at several locations in eastern NC between August 1997 and April 1998. They reported that the data indicate that dry deposition of N as ammonia/ammonium at an eastern farm site was nearly twice that from wet deposition. This data emphasized the highly variable nature of dry deposition and the complex spatial patterns to be expected in an area like eastern NC that is dotted with strong sources (9).

Whitall and Paerl (2001) reported on a modeling effort done by the USEPA at two CASTNet (Clean Air Status and Trends Network) stations which estimated the dry deposition of particulate and gaseous inorganic N in eastern NC. They indicated that the concentrations reported would represent up to a 33% increase over the Neuse River watershed-wide average N deposition. They also pointed out that the techniques used to estimate the dry deposition may have underestimated the actual deposition, meaning that the dry depositional flux may be equal in magnitude to the wet depositional flux in certain areas of the coastal plain (3).

Neuse Basin Specific Research Information

Several studies attempted to assess the direct and indirect contribution from wet atmospheric N deposition to the Neuse River Basin and ultimately the additional contribution of nitrogen to the Neuse River Estuary and the Pamlico Sound.

Paerl et al. (2002) used a nitrogen retention and an instream degradation model to estimate the amount of indirect N deposition reaching the estuary. This value combined with the measured direct depositional flux demonstrated that the wet atmospheric N contribution is between 15% and 31% of the new or externally supplied N flux to the estuary on an annual basis with direct deposition to the estuary alone accounting for 5% of the total new N flux (1). They found that the deposition rates varied seasonally with the highest fluxes occurring during the summer months, which may be driven by seasonal changes in emissions (1). They also noted that downstream of the Neuse Mesohaline (referring to estuarine water with a salinity ranging between 5-18 ppt) N stripping zone, atmospherically deposited N may represent as much as 40% of new N inputs to the open Pamlico Sound (1).

Whitall et al. (2003) attempted to assess the importance of atmospheric deposition to the annual nitrogen budget of the Neuse River Estuary (based on data collected between July 1996 and July 2000). They reported spatial variability in the annual wet N atmospheric deposition with the highest flux occurring in the middle segment of the watershed and the lowest fluxes occurring in the upper watershed (2). They reported the total wet atmospheric N deposition to the land area of the Neuse River basin to be approximately 16.9 Gg N/yr (2). They estimated the total N flux from all sources to the estuary to be 7.5 Gg N/yr and, of this, the wet atmospheric N deposition for direct (at the estuary = 0.4 Gg

N/yr) and indirect (0.7 to 3.7 Gg N/yr) deposition to account for between 1.1 and 4.1 Gg N/yr to the estuary. Thus, the total atmospheric fluxes represent between 15% and 55% of the total new N flux to the estuary, while the direct deposition accounted for about 5% of the new N flux (2). In reviewing the NADP annual NH_4^+ deposition graph for the Sampson County Station (see Figure 3), the deposition is continuing to rise and is likely that the wet and dry atmospheric N deposition contribution will continue to increase in the Neuse as well. Most of the studies reported here did not address the dry deposition of nitrogen which may be a significant additional flux of N to the estuarine system (2). See Table 3 for corresponding nitrogen numbers in pounds per year to the estuary and the entire land area in the Neuse River Basin.

Table 3. Whitall et al. (2003) (4 year dataset) corresponding nitrogen numbers in pounds per year (1Gg = 2,204,622.6218488 lbs) (2).

	Nitrogen in (lbs/yr)
Total (from all sources) N flux to the estuary.	16,534,669
Atmospheric* N deposition to land areas in the Neuse Basin.	37,258,122
Direct atmospheric* N deposition to the estuary.	881,849
Estimated estuarine flux of indirect & direct atmospheric* N deposition.	2,425,084 – 9,033,952

* Wet atmospheric N deposition only.

Whitall and Paerl (2001) estimated that the direct wet atmospheric N deposition accounted for up to 4% to 6% of the total new N loading to the estuary (based on data collected between July 1996 and July 1999). They also discussed the importance of understanding the potential contribution from direct atmospheric N deposition when assessing the new N budget for the Pamlico Sound due to its large surface area (4500 km2) (3). They concluded that the wet atmospheric N deposition is an important component of the N flux to the Neuse River Estuary, accounting for approximately 24% (a range of 15-51%) of the total new N flux to the estuary and that these fluxes vary seasonally and spatially within the watershed (3). See Table 4 for nitrogen contribution in pounds per year to the estuary.

Table 4. Whitall and Paerl (2001) (3 year dataset) nitrogen in pounds per year (3).

	Nitrogen in (lbs/yr)
Total (from all sources) N flux to the estuary.	16,126,814
Direct atmospheric* N deposition to the estuary.	848,559
Best estimate (24%) of indirect atmospheric* N deposition to the estuary (range 15-51%).	3,112,927 (range 1,582,478 – 7,403,563)

* Wet atmospheric N deposition only.

The DAQ 1999 "Status Report on Emissions and Deposition of Atmospheric Nitrogen Compounds from Animal Production in NC", reported that the proportion of total N that enters these coastal waters from the atmosphere varies substantially and for the Albemarle-Pamlico Estuary it ranges between 38% and 44% of the total N (9). DAQ also reported that the USEPA estimates that more than 40% of the nitrogen entering the Albemarle-Pamlico Sound comes from the atmosphere. (9)

Conclusion

It is clear that atmospheric nitrogen deposition, wet and dry, is an important component of the total nitrogen flux to the Neuse River watershed and is likely a significant contributor to the over eutrophication and productivity of the Neuse River Estuary. It also appears that the intensive animal production in the eastern region of the state is influencing the direct and indirect nitrogen inputs to the coastal ecosystem. Some form of controls on the release of these nitrogen compounds might be required before attainment of the state standard in the estuary is feasible.

While continued reduction from other point and nonpoint sources of nitrogen in the watershed need to be sought through strengthening stormwater rules and the permitting processes, attainment of the 30% reduction in nitrogen load may be difficult to achieve without addressing atmospheric contributions to the watershed. Research to date indicates that atmospheric contribution accounts for 15 to 55% of the total nitrogen to the Neuse River Estuary and that these contributions have risen over the last two decades with the increase in concentrated animal operation in the coastal region of our state.

While a moratorium on new hog farms occurred in 1997, other high density animal operations have continued to grow in the area (broilers, layers, and turkey) which are also known to contribute large amounts nitrogen to the atmosphere (15). In 2007, Senate Bill 1465 was signed into law prohibiting the construction of new hog waste lagoons and sprayfields as the primary method of waste management. This law also assists farms to voluntarily convert their current waste system to new innovative animal waste management systems described in the law as well as established a Swine Farm Methane Capture Pilot Program.

Under this new law, new or expanding farms will be allowed as long as they use the new waste technologies, which require the elimination of animal waste discharge to surface water and groundwater through direct discharge, seepage, or runoff and the substantial elimination of atmospheric emission of ammonia. It is currently unknown how this new law and these new waste management technologies will affect the atmospheric emissions from hog facilities. Existing hog farms are not required under this law to convert to using the newer more environmentally friendly waste management system. All conversions are voluntary at this time. It is important that the siting of new farms take into account the regional water quality, current atmospheric nitrogen concentration, if there are N-sensitive waterbodies downwind as well as avoid placement within a flood plain and areas with sandy soils and shallow groundwater (16).

Additional research is needed in order to get a complete understanding of the magnitude and variability of all nitrogen inputs into the system. An understanding of these is necessary to improve basinwide water quality management. The current management strategy attempts to control nitrogen from sources that are better understood because of supporting data history and hence are more tangibly manageable. Atmospheric nitrogen deposition does contribute to the nitrogen loading in the state's waterways but is not managed because of the lack of knowledge on controlling emissions. A commitment is needed now to decrease contributions from atmospheric nitrogen sources, which can be adjusted to reflect new technologies and data as they become available.

Management Information

The following are some of the management suggestions taken directly from the papers reviewed:

Management of N emissions, transport, and deposition must be incorporated in air/watershed and larger-scale nutrient management schemes (1).

It would appear very difficult for a watershed or state (due to airshed size) to have a major effect on atmospheric N deposition if acting alone. Efforts to address coastal atmospheric N deposition management need to be as large as the multi-state regional air pollution program for ozone and fine particulates spawned by the 1990 Clean Air Act Amendments to be efficient and effective (1).

Develop a comprehensive strategy for reducing the nitrogen problems (4).

1. Develop more effective and broadly applied controls on nitrogen oxide emissions from internal combustion engines in automobiles, boats and all terrain vehicles, lawnmowers, chain saws, and other fossil fuel-powered tools and machines.
2. Develop more effective and broadly applied nitrogen oxide controls on industrial and power plant emissions (i.e. stack emission controls).
3. Minimize open-air storage of animal waste and other reduced nitrogen product and sources and improve treatment of animal wastes using on-site "treatment plants" and engineered wetlands. Use recycled water in animal operations to minimize the generation and storage of liquid animal waste.
4. Recycle accumulated solid waste into commercial fertilizers. Apply nitrate, ammonium and urea-based fertilizers at agronomic rates.
5. Use "controlled" burns to minimize atmospheric "fertilization" of downwind nitrogen-sensitive waters with either nitrogen oxides or ammonia/ammonium.

Though limited in spatial extent, high deposition rates near the source are likely to exceed the critical nitrogen loads for most ecosystems, suggesting that siting requirements for animal production facilities should consider local nitrogen deposition as a potential environmental burden (6).

Water contamination from CAFOs, from nitrogen, phosphorus, microbes and antibiotics, may increase with concentration of livestock and liquid manure storage and spreading. Contamination risk may decrease with less livestock concentration, proper waste management and well planned CAFO siting that considers regional water quality and avoids sandy soils, shallow groundwater and flood plains (16).

Future Research

In 1996 the NC legislature appropriated $450,000. The appropriation was assigned to the UNC board of Governors for research efforts (NCSU administered these funds to researchers in NC). The primary purpose of the funding was for modeling of the emissions, fate and deposition of nitrogen compounds. EPA also committed $500,000 in support of the modeling effort. (9) Much of the research reviewed here may have resulted from this effort by the legislature to increase our understanding of atmospheric deposition.

The USEPA and the USDA jointly asked the Board of Agriculture and Natural Resources to evaluate the scientific information needed to address several issues such as the need for regulations of air emissions from animal feed operations (AFOs) and how to mitigate the effects of air emission with modified agricultural practices. The board concluded that there was a lot of research needed before these types of questions could be answered. A special study is currently underway by the EPA to address the concerns found. This study will improve the estimates of emissions from the different sources of AFOs. Monitoring was to start in the spring of 2007. Twenty five sites at 21 farms were chosen and are located in 10 different states. Three sites are located in NC (2 swine and 1 poultry). Selected sites will be monitored for 2 yrs to ensure that the data account for seasonal variability as well as the impact of any operational changes. Monitoring will take place at barns, lagoons and waste or manure storage piles. (17)

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