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Atmospheric nitrogen compounds II: emissions, transport, transformation, deposition and assessment

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Abstract

The Atmospheric Nitrogen Compounds II: Emissions, Transport, Transformation, Deposition and Assessment workshop was held in Chapel Hill, NC from 7 to 9 June 1999. This international conference, which served as a follow-up to the workshop held in March 1997, was sponsored by: North Carolina Department of Environment and Natural Resources; North Carolina Department of Health and Human Services, North Carolina Office of the State Health Director; Mid-Atlantic Regional Air Management Association; North Carolina Water Resources Research Institute; Air and Waste Management Association, RTP Chapter; the US Environmental Protection Agency and the North Carolina State University (College of Physical and Mathematical Sciences, and North Carolina Agricultural Research Service). The workshop was structured as an open forum at which scientists, policy makers, industry representatives and others could freely share current knowledge and ideas, and included international perspectives. The workshop commenced with international perspectives from the United States, Canada, United Kingdom, the Netherlands, and Denmark. This article summarizes the findings of the workshop and articulates future research needs and ways to address nitrogen/ammonia from intensively managed animal agriculture. The need for developing sustainable solutions for managing the animal waste problem is vital for shaping the future of North Carolina. As part of that process, all aspects of environmental issues (air, water, soil) must be addressed as part of a comprehensive and long-term strategy. There is an urgent need for North Carolina policy makers to create a new, independent organization that will build consensus and mobilize resources to find technologically and economically feasible solutions to this aspect of the animal waste problem.

Keywords: Ammonia; Nitrogen compounds; Emissions; Effects; Transport; Transformation; Swine operations and abatement

1. Background

Nitrogen is perhaps the most important nutrient governing the growth and reproduction of living organisms. Nitrogen compound emissions also have a profound effect on air quality. Two major needs that drive the contemporary perturbations of the nitrogen cycle are the seemingly insatiable human appetite for energy, leading to the emission of nitrogen oxides into the atmosphere, and the need for food to sustain growing numbers of people all over the world, leading to the agricultural emission of ammonia. Once released into the atmosphere by either man-made (anthropogenic) or natural sources,

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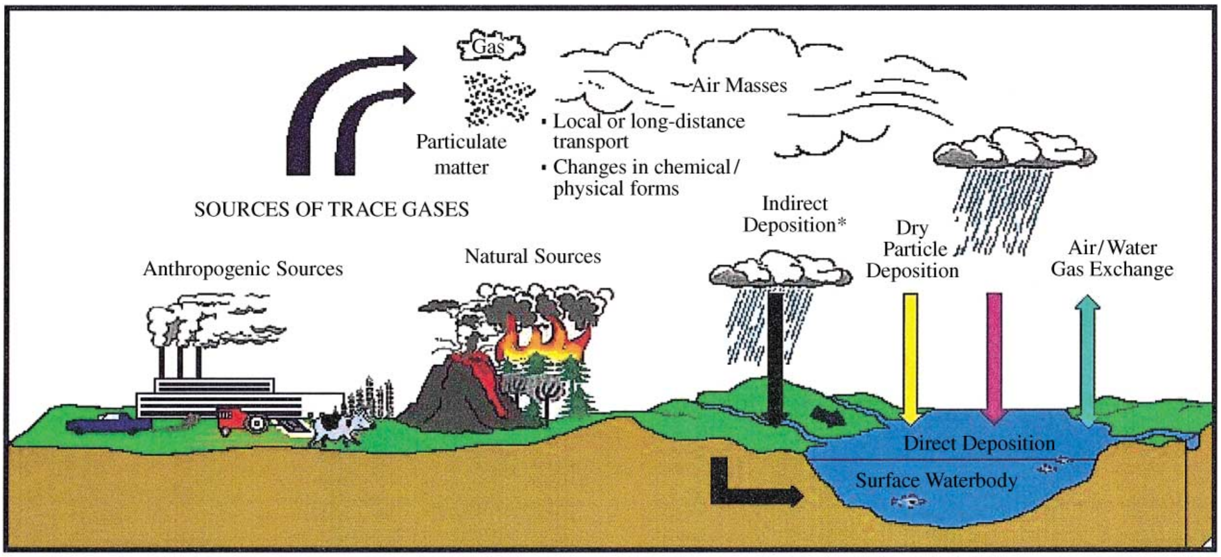

* Indirect deposition is direct deposition to land followed by runoff or seepage through groundwater to a surface waterbody.

Fig. 1. Atmospheric emissions, transport, transformation and deposition of trace gases.

these nitrogen compounds can undergo several different processes such as transformation due to atmospheric reactions (e.g. gas-to-particle conversion), transport associated with wind, and finally wet and dry deposition (Fig. 1). All of these processes can perturb the environment with a host of beneficial and detrimental effects, such as increased crop yields from nitrogen loading or decreased visibility from increased aerosol production. Table 1 represents the current global estimates for sources and sinks of several key nitrogen species (oxidized nitrogen compounds, nitrous oxide, and ammonia). Scientists have focused recently on the oxidized species of nitrogen ($NO_x = NO + NO_2$) and their role as precursors to ozone (O_3) formation, and the reduced species (NH_x = ammonia + ammonium + amines) and their role in nitrogen enrichment and eutrophication of aquatic ecosystems. Nitrous oxide (N_2O), while contributing to ozone destruction in the stratosphere, is relatively inert in the troposphere and therefore has negligible consequences in tropospheric photochemistry, but does contribute to climate change as a greenhouse gas (Warneck, 1988).

2. Emissions

Fossil fuel combustion has increased to meet growing energy demands. The global amount of fossil fuel use per person (Fig. 2) has increased by more than a factor of 6 over the last 75 years. At the same time, scientists have synthesized nitrogen-based fertilizers to enhance crop development and to maximize production on limited land space. Before the mass production of fertilizers, it can be assumed that there was an approximate balance between the relatively unreactive molecular nitrogen (N_2 comprises approximately 80% of air) in the atmosphere, which was naturally converted to forms used by plants and animals, and the amount of nitrogen returned to the atmosphere via natural processes (Delwiche, 1970). Currently, however, the global production of fertilizer is approximately 100 million metric tons of nitrogen yr^{-1}, compared to approximately one million metric tons only 40 years ago (The Fertilizer Institute, 2000). The results of increased fertilizer and power production have reached a point where the scientific community has major concerns about the fate of the nitrogen produced.

Estimates of NH_3 emissions and the contribution from different source categories given in Figs. 3 and 4 show that hog operations are responsible for a larger percentage of the nitrogen budget in North Carolina than they are in the US as a whole. The relatively large NH_3 contribution from hog operations in North Carolina as compared to the US as a whole can be explained by Fig. 5 which shows the growth of the hog industry during the last two decades. Data presented at the workshop (Fig. 6) revealed that NH_3 emissions in a six-county (Bladen, Duplin, Greene, Lenoir, Sampson, Wayne) area of North Carolina that maintains the state's densest and largest population of hogs (Fig. 7) increased significantly during the same time period that the hog operations increased (Walker et al., 2000). Mean NH_3 emissions from hog operations increased 316% between 1982–1989 and 1990–1997; 84% of the growth from all sources (i.e., hogs, fertilizer, cattle, turkeys, broilers, chickens) can be

Table 1
Global atmospheric budgets of NO_x, N_2O, and NH_3

Source or sink	NO_x [a]	N_2O [b]	NH_3 [c]
	(Tg N yr^{-1})[d]		
Fossil fuel combustion	21	0.5	2
Biomass burning	8.0	0.4	5
Sea surface	< 1.0	5.7	13
Domestic animal waste	—[e]	1.6	32
Human excrement	—	—	4
Lightning	8	—	—
NH_3 oxidation by OH	1	0.6	—
Stratospheric input	0.5	—	—
Soil emissions	20.2	10.7	19
Other[f]		6.3	
Total sources[g]	59	26	75
Wet deposition	12–42	—	46
Dry deposition	12–22	—	10
Stratospheric sink	—	19.3	—
NH_3 oxidation by OH	—	—	1
Atmospheric accumulation	—	3.5	—
Total sinks	59	19.3	57

[a] Source: Levine (1991).
[b] Source: Bouwman et al. (1995); stratospheric sink from Houghton et al. (1995).
[c] Source: Schlesinger and Hartley (1992).
[d] ($1\,Tg = 10^{12}\,g$).
[e] (—) indicates insignificant or unavailable terms.
[f] Includes adipic and nitric acid production, nitrogen fertilizer, land use change and other small sources.
[g] It is accepted that wet and dry NO_x deposition should total the sum of NO_x sources and that the apparent difference between total NH_3 sources and sinks represents uncertainties in identified budget terms, not atmospheric accumulation.

attributed to the increase in number of hogs (Walker et al., 2000). Fig. 6 also shows that the ammonium ion concentration $[NH_4^+]$ in precipitation collected at a deposition sampling site in Sampson County also increased throughout this period.

3. Atmospheric behavior

Atmospheric ammonia (NH_3) emissions have garnered increased interest in the past few years, due in part to the detrimental effects of excess nitrogen deposition to nutrient sensitive ecosystems (Aneja et al., 1998; Nihlgard, 1985; van Breemen, 1982). Moreover, NH_3 is the most prevalent gaseous base found in the atmosphere, and is, therefore, fundamental in determining the overall acidity of precipitation (Warneck, 1988), cloudwater (Li and Aneja, 1992), and atmospheric aerosols (Lefer et al., 1999). New gaseous ammonia instruments for monitoring and research are currently in advanced stages of development (Erisman et al., 1999). The ecological impact of atmospheric NH_3 deposition may be substantial as reduced nitrogen species are thought to be the most biologically available of nitrogen species in N-limited coastal and estuarine ecosystems (Paerl, 1997). In the atmosphere, NH_3 reacts primarily with acidic species to form ammonium sulfate, ammonium nitrate or ammonium chloride, or it may be deposited to the earth's surface by either dry or wet deposition processes.

The spatial scale of a particular NH_3 source's contribution to atmospheric nitrogen deposition is governed in part by the gas-to-particle conversion rate of NH_3 to NH_4^+. Because of the short lifetime of NH_3 in the atmosphere ($\tau = 1$–5 days or less) (Warneck, 1988), low source height, and relatively high dry deposition velocity (Asman and van Jaarsveld, 1992), a substantial fraction (20–40%) will likely deposit near its source. However, ammonium (NH_4^+) aerosols, with atmospheric lifetimes on the order of $\tau = 1$–15 days (Aneja and Murray, 1998; Aneja et al., 2000) will tend to deposit at larger distances downwind of sources. Ammonia emissions from animal operations contribute substantially to atmospheric nitrogen loading and may contribute the same order of magnitude as emissions of NO in some parts of the world (Steingröver and Boxman, 1996); highlighting the need for new sustainable technologies for intensively managed animal production.

4. Effects

Although nitrogen is a critical nutrient for the survival of micro-organisms, plants, humans and animals, it can cause detrimental effects when concentrations reach excessive levels (Paerl, 1997; Erisman et al., 1998). Fig. 8 (Gundersen, 1992) illustrates this point by showing how an ecosystem responds to increased N loadings. The horizontal line is a crop which receives no atmospheric N deposition, and as indicated by the vertical axis, has a stable index of productivity. However, as N is initially added to the system, the index of productivity steadily increases to the point of diminishing returns, where any additional N loading actually reduces productivity (Schlesinger, 1997). In addition to the productivity concerns of aquatic and terrestrial ecosystems, oxidized and reduced N compounds each play a specialized role in degrading human health and its welfare. Some of the consequences associated with elevated concentrations and depositions of both oxidized and reduced N species are:

1. Respiratory disease caused by exposures to high concentrations of:
   1.1. Tropospheric ozone.

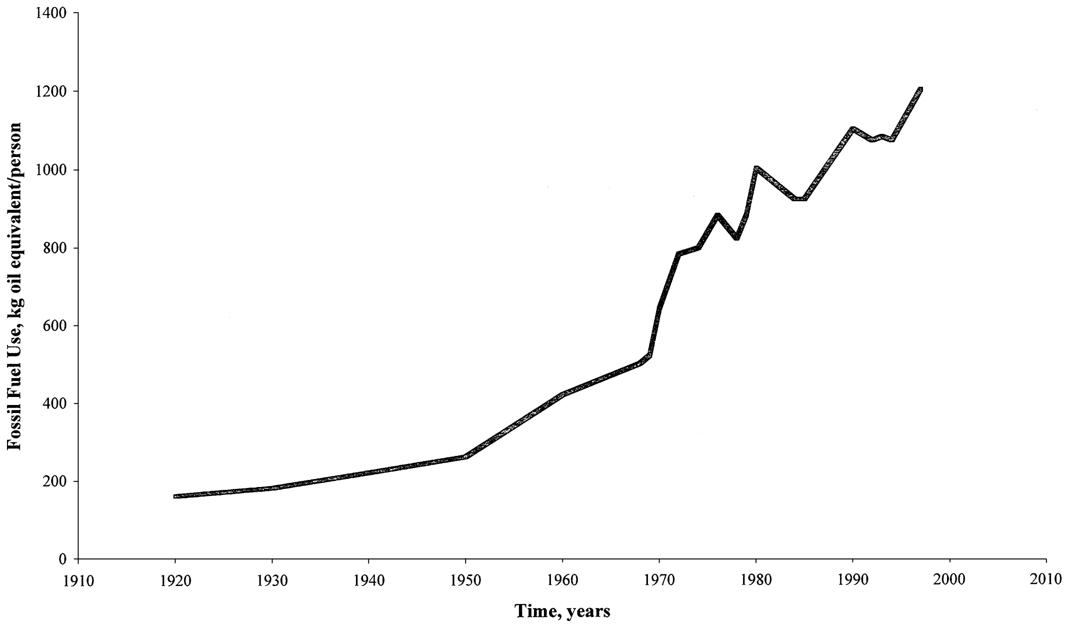

Fig. 2. Trends in global fossil fuel use per person (source: Galloway, 1988).

 - Other photochemical oxidants.
 - Fine particulate aerosol (e.g., PM 2.5).
 - Direct toxicity of NO_2 (on rare occasions).
2. Nitrate contamination of drinking water.
3. Eutrophication, harmful algal blooms and decreased surface water quality.
4. Climatic changes associated with increases in nitrous oxide (greenhouse gas).
5. Nitrogen saturation of forest soils (Erisman et al., 1998).

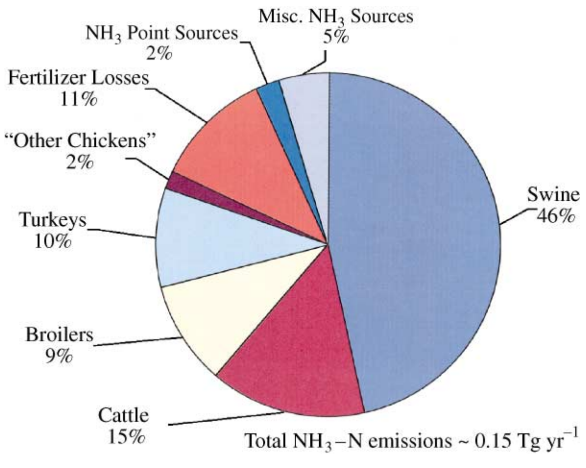

Fig. 3. Percent of ammonia–nitrogen from various sources in North Carolina for 1996 (source: Aneja et al., 1998a).

5. Abatement

Air quality issues associated with intensively managed animal agriculture are now being addressed in Europe and Canada under several initiatives and in consultation and partnership with stakeholders. Emission inventories for several different pollutants including atmospheric nitrogen compounds are maintained by federal governments. For example, the new Air Pollution Protocol for Europe has set reduction targets to be achieved by 2010

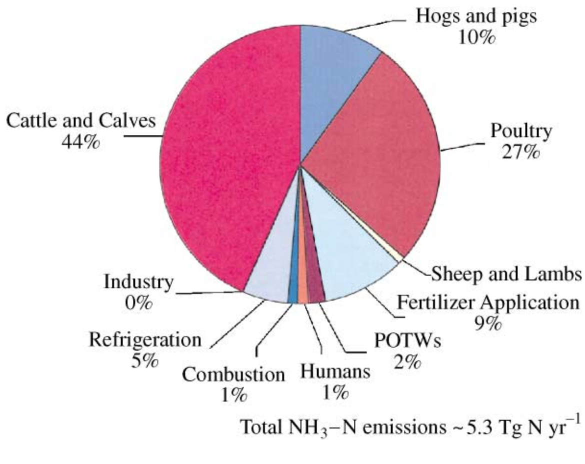

Fig. 4. Relative contribution of ammonia–nitrogen emissions in the US from different source categories (source: Battye et al., 1994).

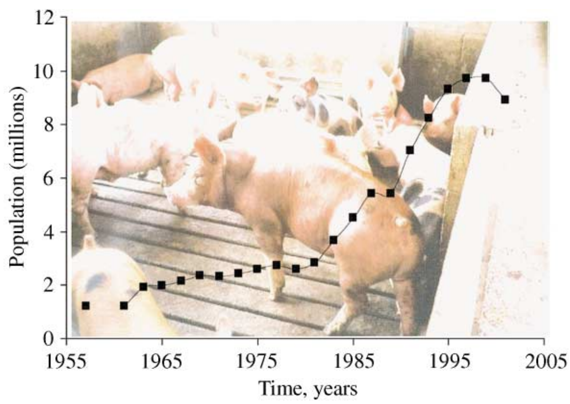

Fig. 5. Hog population in North Carolina (source: North Carolina Agricultural Statistics, 2000).

as compared to 1980 emission levels, for the following: $SO_2 = 63\%$, $NO_x = 41\%$, $VOC = 40\%$, $NH_3 = 17\%$ (http:www.unece.org/press/99env11e.htm). Moreover, in Europe the focus of environmental effects related research is primarily on acidification, eutrophication, biodiversity and groundwater pollution, and the use of critical loads to the ecosystem which accounts for atmospheric deposition pathways.

A "Livestock Environmental Initiative" was launched in Canada during December 1999 in which the livestock industry is working in partnership with the federal government to address environmental concerns through research and development of technology, and for acceleration of technology assessment and transfer. Air quality, including greenhouse gases, is a priority area of concern. Experts from a broad cross-section of government, business and industry, the academic community, environmental groups and non-government organizations have evaluated available information. These options will be reviewed and analyzed to determine the actions needed to reduce emissions.

Improvements in air quality from implementation of the Clean Air Act Amendments (CAAA) of 1990 or other efforts (e.g., Southern Appalachian Mountain Initiative, SAMI) are likely to receive widespread attention only if a target pollutant in question is regulated under the CAAA. At this time, emissions of atmospheric ammonia, ammonium, and organic nitrogen compounds (N_{org}) are not federally regulated, thus minimizing the benefits that might result from the CAAA. In North Carolina, under state law, ammonia is regulated as a toxic air pollutant (15 NCAC 2D.1104(a)(4)).

6. Research needs

The workshop highlighted areas which require further research in North Carolina and elsewhere, such as the further refinement of emission estimates, the role of ammonia and factors that contribute to gas to particle conversion processes (PM_{fine}) in rural/urban and regional areas, computer models to quantify and simulate impacts of deposition, and the establishment of a full scale and continuing monitoring program. Results of the RADM (Regional Acid Deposition Model) and progress that has been made with adapting this model to ammonia deposition were presented at the conference. However, if the main processes and characteristics, specifically

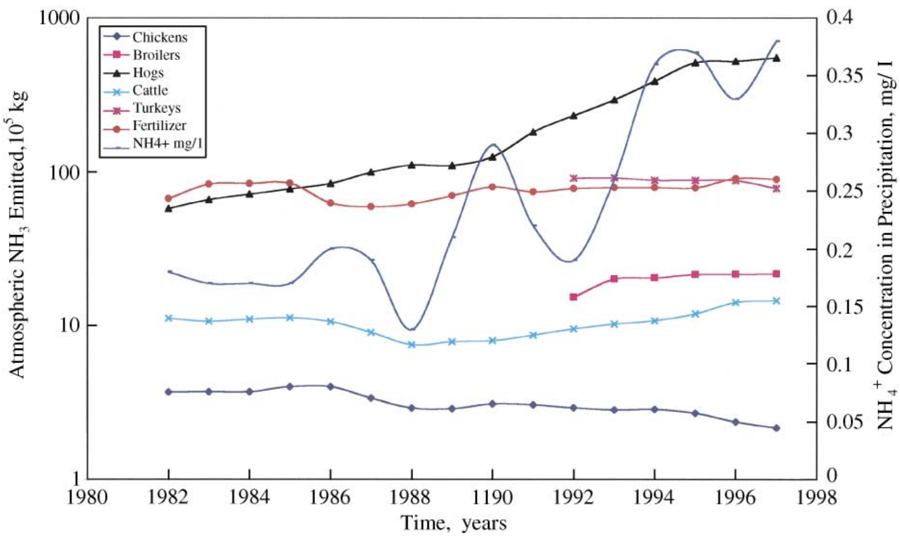

Fig. 6. NH_3 emission estimates by source type in the six North Carolina counties (Bladen, Duplin, Greene, Lenoir, Sampson, Wayne), and annual volume-weighted NH_4^+ concentration in precipitation at Sampson County, North Carolina (source: Walker et al., 2000).

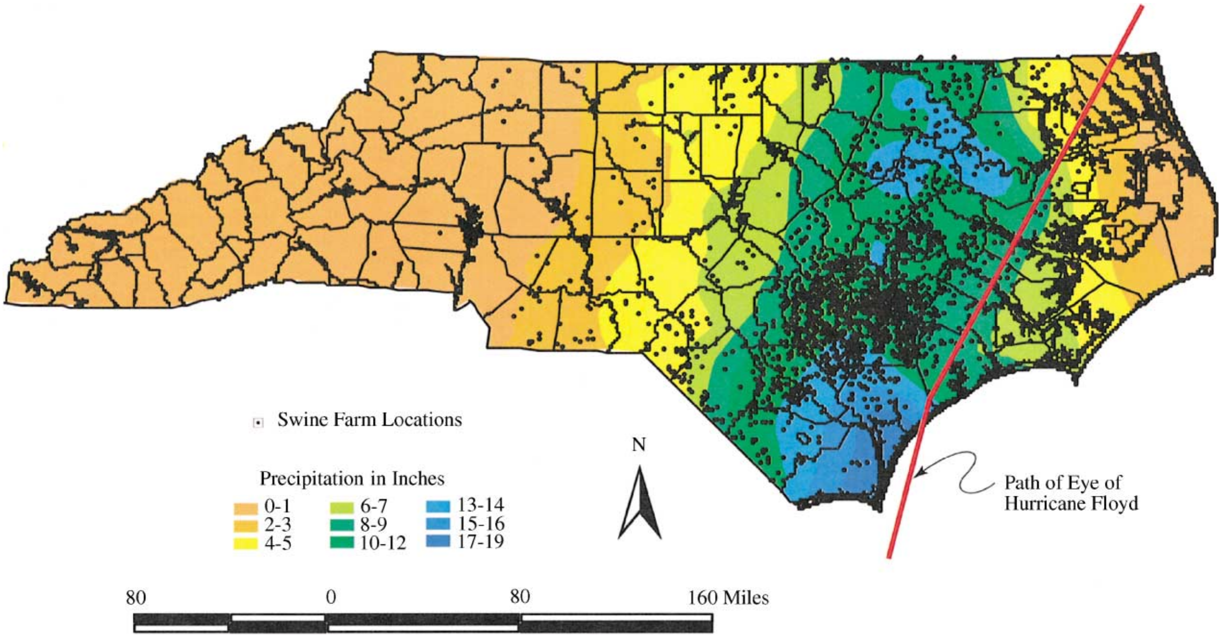

Fig. 7. Map of North Carolina indicating hog sites, rainfall totals associated with Hurricane Floyd (14–16 September 1999), and track of storm.

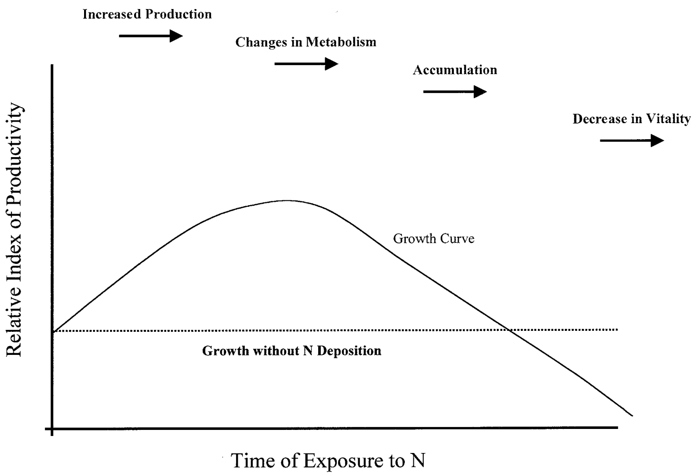

Fig. 8. Hypothetical growth curve for an ecosystem, given different lengths of exposure to nitrogen (source: Gundersen, 1992).

concerning dry deposition, are to be described, then the current grid scaling (20 × 20 km) of this model is still too coarse. Further, a targeted monitoring program in North Carolina needs to be established which includes emissions and both wet and dry deposition at several different land use types. The data collected during this program can then be used to support the modeling effort and assess its performance. A monitoring program will also assist in evaluating any future regulatory policy, which still remains one of the most complicated issues facing North Carolina and the Nation today. Additional research needs are:

1. A detailed understanding of the cycling of atmospheric reduced and oxidized nitrogen compounds, their linkage with emissions of sulphur dioxide (SO_2) and volatile organic compounds (VOCs), and subsequent oxidation products, their spatial and temporal distributions, and their contribution to the chemical composition of aerosols.

Table 2
A comparison of physical and chemical characteristics among the US, North Carolina, and the Netherlands[a]

Parameter	United States	North Carolina	Netherlands
Total land and water area	9,629,000 km^2	136,000 km^2	42,000 km^2
Land area	9,159,000 km^2	126,000 km^2	34,000 km^2
NC coastal plain land area (where majority of hog operations are located)		45,333 km^2	
Inland water area	470,000 km^2	10,000 km^2	6,000 km^2
People	270,312,000 30 km^{-2}	7,651,000 61 km^{-2}	15,731,000 463 km^{-2}
Swine (1996)	56,124,000	9,300,000	14,400,000
Total cattle (1996)	101,656,000	1,100,000	4,412,000
Income from animal agriculture	92.4 billion dollars yr^{-1}	5.7 billion dollars yr^{-1}	4.4 billion dollars yr^{-1}
1995 NO_x emissions	21,600,000,000 NO_x yr^{-1} 6,560,000,000 kgN yr^{-1}	570,000,000 kgNO_x yr^{-1} 173,000,000 kgN yr^{-1}	518,000,000 kgNO_x yr^{-1} 158,000,000 kgN yr^{-1}
1995 NH_3 emissions	2,730,000,000 kgNH_3 yr^{-1} 2,250,000,000 kgN yr^{-1}	155,000,000 kgNH_3 yr^{-1} 127,000,000 kgN yr^{-1}	152,000,000 kgNH_3 yr^{-1} 125,000,000 kgN yr^{-1}

[a]Source: http://www.cia.gov. http://www.minlnv.nl/international/stat/factagricult1.htm. http://www.usda.gov/news/pubs/fbook98/ch3g.htm. http://www.epa.gov/ttn/chief/trends97/browse.html.

2. Need to know the contribution of atmospheric deposition of ammonia/ammonium to estuarine and coastal N loading.
3. Need to better understand the ecological effects of ammonia/ammonium as a new N source causing eutrophication of N-sensitive waters.

7. Summary and conclusions

Although North Carolina faces many challenges regarding nitrogen issues, the problem is not limited to North Carolina. Many of the issues which face politicians, farmers, citizens, and international researchers are similar. Therefore, much can be gained through collaboration and exchange of ideas. For example, a comparison between various factors which influence the emission and deposition of total fixed nitrogen (Table 2) in the Netherlands and the US reveals striking similarities. Although land size and human and animal populations differ, the estimates for NO_x and NH_3 emissions, income from agriculture, and inland water areas (adjusted for coastal districts in NC) are all very similar (Table 2). Due to the many similarities and the fact that North Carolina's rapid growth in animal husbandry started almost 2 decades later than the Netherlands, North Carolina can significantly benefit from their experiences.

The current technology used in North Carolina to manage the hog waste is known as the Lagoon and Spray System, which consists of an exposed waste lagoon to store the waste (~98% liquid) and mechanisms through which the waste is periodically sprayed onto the crops as a nutrient source. The technology can be subdivided into four distinct processes (Fig. 9), all of which release NH_3 to the atmosphere: Production houses; Waste Storage and Treatment Systems (Aneja et al., 2000) (Fig. 10); Land application i.e., spraying; and Biogenic Emissions

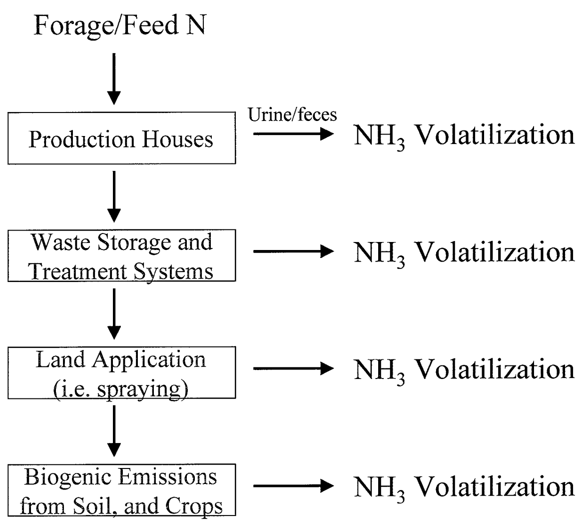

Fig. 9. Major routes for NH_3 emissions from intensively managed animal operations in North Carolina, USA.

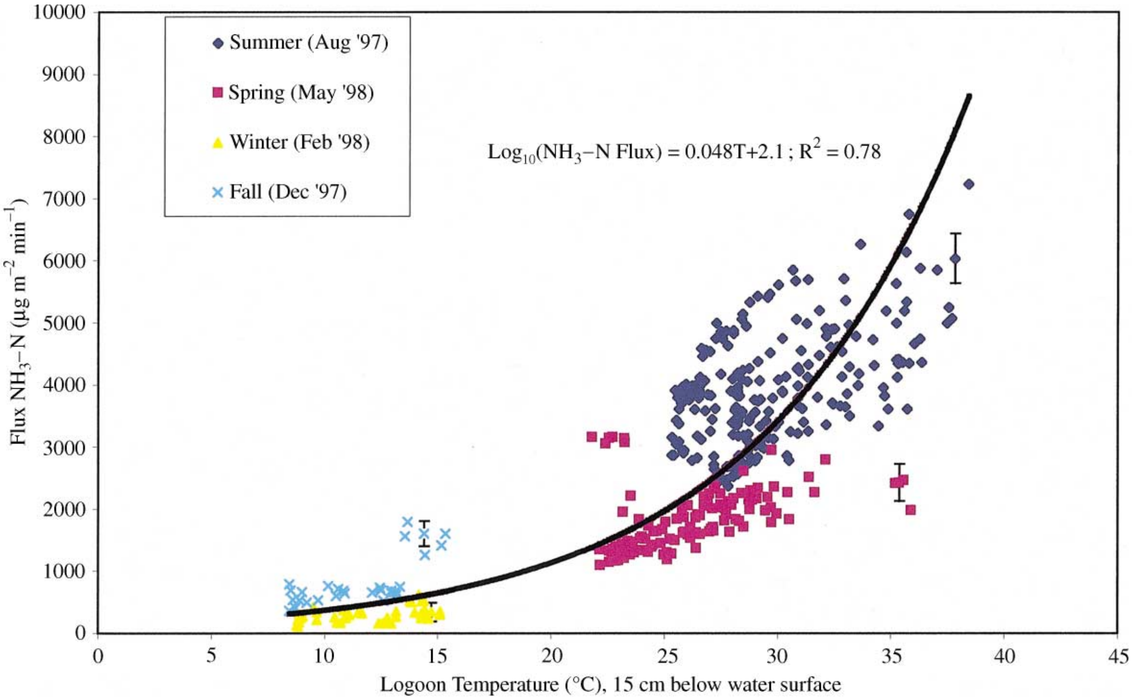

Fig. 10. North Carolina lagoon ammonia–N flux versus lagoon surface water temperature. pH of lagoon: 7–8 pH units, total Kjeldahl nitrogen (TKN) in lagoon: 500–750 mg-N^{-1}. Vertical bars represent one standard deviation (source: Aneja et al., 2000).

from Soil and Crops. Current estimates of NH_3 emissions in North Carolina from hogs alone, utilizing an emission factor (20.3 lb of NH_3 hog^{-1} yr^{-1}) determined by Battye et al. (1994) are: 1994, ~ 195 t of NH_3 d^{-1}; 1996, ~ 258 t of NH_3 d^{-1}; 1999, ~ 264 t of NH_3 d^{-1} (where t = metric tons, and d = day). The lagoon and spray system requires continuous attention due to its susceptibility to flooding, the potential for release of waste to nearby water sources, and also due to odor issues.

The lagoon system recently gained renewed national attention in the aftermath of Hurricane Floyd (15–16 September 1999). The eye of the storm passed over the most intensively managed animal husbandry sites in North Carolina (Fig. 7). The storm resulted in the death of approximately 3 million chickens and turkeys, 880 cattle and 30,000 hogs with many of the carcasses floating in the flood waters; 50 animal operations with waste lagoons were flooded, allowing millions of gallons of animal waste to be spilled into flood waters; and 24 municipal wastewater treatment plants were flooded (WRRI News, 1999). The environmental consequences of this disaster, not yet fully known, include nitrogen release from lagoons and wastewater treatment plants.

Sustainable solutions must be found for managing the animal waste problem in North Carolina. As part of that process, all aspects of environmental issues (air, water, soil) must be addressed as part of a comprehensive and long-term strategy. There is an urgent need for North Carolina policy makers to create a new, independent organization that will build consensus and mobilize resources to find technologically and economically feasible solutions to this aspect of the animal waste problem.

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