

Project Title: Changes in Ammonia Emissions from North Carolina Swine Lagoons Associated with Improved Production Management

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Industry Summary: Swine production and manure management and storage have been implicated as a major and increasing source of ammonia (NH₃) emissions in North Carolina, with increasing environmental impact. Further, reports have stated that ammonium (NH₄⁺) deposition, as measured by the National Atmospheric Deposition Program (NADP, a program established to determine long-term trends in NH₄⁺ and other nutrient deposition in the United States), increased about 100% during periods overlapping the swine-production expansion period (1990-1996) in North Carolina, without considering deposition trends before or after the industry expansion or other changes that would have contributed to NH₄⁺ depositions. As a result of this and other concerns, the North Carolina legislature enacted a moratorium on new swine farms in 1997. The swine industry questions this conjecture. Because of a lack of emissions data, the United States Environmental Protection Agency (USEPA) initiated studies across North Carolina to determine NH₃ emissions from livestock operations under a USEPA Air Consent Agreement with the livestock industries. These studies were undertaken from 2007 to 2009, as a National Air Emissions Monitoring Study (NAEMS) funded by participating producers. These NAEMS studies estimated that swine NH₃ emissions were 1.5 times larger from finisher and 18.3 times larger from sow production systems over what had been measured by a USDA study 10 years prior. The NAEMS authors suggested their increased estimates were likely caused by different measurement approaches and climate conditions but ignored other factors that affect emissions. The swine industry challenges the accuracy of the reported increase in emissions from swine farms since over this period the industry made numerous changes to improve feed efficiency which should have reduced emissions.

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This study was initiated to establish how long-term enhancements in animal management have affected nutrient loading in lagoons. Regular lagoon sampling for chemical analysis has been required for lagoon permitting in North Carolina and the data from swine facilities have been archived for about 20 years. To evaluate the representativeness of the archived data, simultaneous lagoon sampling by farm employees and by the authors were performed to compare source and magnitude of sampling, analytical technique, and potential sample spatial variability. The simultaneous sampling and analysis showed no statistical difference between farm employee- and researcher-collected data, assuring the archived data's usefulness in estimating trends in NH₃ emissions, based on lagoon's nutrient contents and climatic conditions. Archived results from samples taken on 159 primary waste-processing lagoons along with 23 secondary lagoons (not all farms had secondary lagoons), and representing about 106,000 laboratory analyses, were used for analysis.

Nutrient content trends in finisher primary lagoons measured excreted nutrient concentration reductions [total Kjeldahl nitrogen (TKN), phosphorous (P), potassium (K), and zinc (Zn)] ranging from 35% to 78% in the primary lagoons, with the exception of copper (Cu) which increased by 41% (Copper is an essential dietary nutrient and supplementation above the minimum nutrient requirement can be used as an alternative to antibiotics for improving growth and feed efficiency at finisher farms). All primary lagoon nutrient trends for sow farms, including Cu, also declined ranging from 17% to 68%. Nutrients in all secondary lagoons similarly declined, ranging from 27% to 95% decrease as the industry implemented swine management improvements. The trends in primary lagoon pH for finisher and sow farms showed decreases of 0.10 and 0.02 units, respectively, and 0.02 units for both types of farms' secondary lagoons. The lagoon N and pH data, along with regional climate data plus process and empirical NH₃ volatilization models, were used to calculate the changes in relative NH₃ emissions over time and to examine the NH₃ emissions' relationship to improved swine production and management practices. Because of reduced N and decreased pH in the lagoons, the USDA process model for NH₃ emissions showed trend decreases since 2001 of 47% and 22% from finishers' and sows' primary lagoons, respectively, and 49% and 50% from finishers' and sows' secondary lagoons, respectively. Additional statistical and empirical models estimated reductions in NH₃ emissions ranging from 11% to 54%, based on the same archived data from all farms' primary and secondary lagoons.

From 1979 through 2017, the period in which NH₄⁺ deposition data is available, the swine population in North Carolina had three time-periods with very different but approximately linear growth rates: pre-1989 (pre-expansion period), 1990 through 1996 (expansion period), and 1997 through 2017 (post-expansion moratorium period). Human population growth in North Carolina during those three time periods was also approximately linear, with growth rates of 75,000 persons yr⁻¹ pre-1989 and about 135,000 persons yr⁻¹ during and after the swine-expansion periods. It might be expected that when the average yearly change in swine production increased from 44,000 swine yr⁻¹ to 1,132,000 swine yr⁻¹ in 1990 and

then after 1996 (the moratorium period) down by a loss of 47,000 swine yr⁻¹, there would be dramatic changes in deposition growth as measured by the NADP; but, this was not observed either statistically or graphically. Overall (1979 through 2017), NH₄⁺ deposition at both rural and urban monitoring sites showed statistically significant linear increases (i.e. deposition approximately increased at a constant rate over time). However, when each separate time period was tested for a linear relationship (correlation), the calculated probabilities (*p*-values) were too large (*p* = 0.18, *p* = 0.11 and *p* = 0.06 respectively) to claim a significant linear relationship between deposition and time. Since more data values would possibly allow for significant correlations, both monthly and weekly data were also evaluated. While the long-term trends had low calculated probabilities (*p* < 0.001), separate period linear relationships were not consistent, with the slopes changing with each data set and time period. However, because of variability in the data, a significant change in deposition during the period of rapid industry expansion cannot be claimed or disproven nor to what might have been the magnitude of possible changes for the pre- and post-expansion periods.

It is noteworthy that a visual inspection of the data shows lack of noticeable change in deposition growth after the expansion period. If swine population was the primary driving force for the increase in deposition during the period of rapid industry expansion, then after the moratorium the decrease in swine population combined with improved swine feed efficiency (-8%) and daily gain (+22%) would have resulted in a slowed growth of deposition in the years post-1996, but this was not observed. And, during this post-expansion period, fecal N and P were reduced by 34% and 62%, respectively. The increase in NH₄⁺ deposition during the post-expansion period is an indicator that non-swine factors affect NH₄⁺ deposition in both rural and urban North Carolina.

In summary, an analysis of archived lagoon nutrient data collected for the swine industry permitting process indicated a significant reduction in the industry's NH₃ emissions' footprint, as well as other nutrients, since initiation of lagoon sampling began in 2001. These data confirm that industry management and production improvements have been effective in reducing elemental excretion. Analysis of NADP data also indicated that an increase in rural and urban NH₄⁺ deposition over the last 40 years, which has been attributed to increasing swine population and other agricultural production, is neither accurate nor complete and that human population and its associated NH₃ emissions must be investigated as a significant factor.

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Scientific Abstract: Swine manure management and storage has been implicated as a major source of increasing agricultural ammonia (NH_3) emissions in North Carolina. This study was conducted to establish how improvements in manure and animal management have affected nutrient loading in lagoons, reduced NH_3 emissions, and had effect on ammonium (NH_4^+) deposition in rural and urban areas. Periodic lagoon sampling for chemical analysis has been required for lagoon permitting and the data from swine facilities has been archived by the state of North Carolina. Archived lagoon chemistry analyses data from 182 farm lagoons were used to evaluate trends in lagoon chemical properties. Process and empirical NH_3 volatilization models were used with the data to calculate the relative changes of NH_3 emissions (since 2001) to improvements in swine production and management practices. Trends of lagoon nutrient content for both finisher and sow farms, primary and secondary lagoons, showed all measured nutrient elements in lagoons decreased from 17% to 95% except for a 41% increase in copper (Cu) in finisher primary lagoons (Copper is an essential dietary nutrient and supplementation above the minimum nutrient requirement can be used as an alternative to antibiotics for improving growth and feed efficiency at finisher farms). Because of reduced N and pH in the lagoons, a process model for NH_3 emissions suggested decreases from primary lagoons of 47% and 22% from both finisher and sow farm lagoons, respectively. Empirical and semi-empirical models predicted even larger decreases in NH_3 emissions ranging from 11% to 54% since 2001, depending on the animal type, primary or secondary lagoon, and emissions' model used. Long-term National Atmospheric Deposition Program (NADP) deposition measurements showed NH_4^+ deposition increased linearly with time at both the rural and urban measurement sites. Our analysis of lagoon nutrient concentrations, NH_3 emissions, and NADP data indicates that the increase in rural and urban NH_4^+ deposition over the last 40 years, which have been largely attributed to increasing swine population, do not reflect changes in swine population or improved production practices, and that human population growth or other NH_3 emission sources must be investigated as significant factors.

Changes in Ammonia Emissions from North Carolina Swine Lagoons
Associated with Improved Production Management

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Introduction:

Swine manure management and storage has been implicated as a major source of atmospheric ammonia (NH₃) emissions in North Carolina (Aneja et al., 2000; Grant et al., 2016). Ammonia from any source may be neutralized by atmospheric concentrations of available acid gases creating particulates (PM_{2.5}) which may be transported as aerosols, resulting that any NH₃ source may become part of an airshed impacting distant ecosystems (Harper et al., 2004). Reports have estimated (Aneja et al., 1997; Aneja et al., 2000; Walker et al., 2000) that nitrogen (N) emissions from swine operations are the largest statewide contributor of NH₃, accounting for almost half of North Carolina's NH₃ emissions and about 20% of all total nitrogen (N) emissions. Animal population numbers, provided by the North Carolina Department of Agriculture and Consumer Services (NCDA&CS, 2019), have been used with the animal emission factors for swine [which were based on a report by Battye et al. (1994) developed mainly from data of Northern Europe] for calculating statewide NH₃ emissions. The data were variable and questionable for use in estimating NH₃ emissions in the southern Coastal Plain of the United States as was suggested by the author of the European data (Asman, 1992) that the emission factors may have been too large by about a factor of two (Harper et al., 2004). Comparison of Battye's emission factor of 9.2 kg NH₃ animal⁻¹ yr⁻¹ for North Carolina, to that of Asman's of 4.4 in Northern Europe, and to emission factors measured by the U.S. Department of Agriculture (USDA) of 1.1 (finisher farms) and 1.7 (sow farms) in North Carolina (Harper et al., 2004), indicates that high NH₃ emissions estimated to be from swine in North Carolina should be considered questionable.

A paucity of measured NH₃ emissions led the United States Environmental Protection Agency (USEPA) to institute studies across the U.S. to determine NH₃ emissions (under a USEPA Air Consent Agreement with the animal industries) as a National Air Emissions Monitoring Study (NAEMS) funded by participating producer assessments, undertaken from 2007 to 2009. The NAEMS emissions were to be used to address concerns over the deposition of ammonium (NH₄⁺), to ensure compliance with regulatory standards, to evaluate production efficiency, and to determine industry trends. To augment emissions information of the USDA 1997-1999 microclimate-technique measurement studies in North Carolina, the NAEMS study evaluated two farms (finisher and sow) also using non-interference microclimate techniques.

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The NAEMS studies in North Carolina found emission factors from the swine industry that were considerably larger than those found 10 years prior by the USDA study (Harper et al., 2004; Harper and Sharpe, 1998). The USDA study, using complete-day (daytime plus nighttime) measurements over two years, determined average annual emission rates of 3.0 and 4.7 g NH₃ an⁻¹ d⁻¹ from breed-to-finish and sow lagoons, respectively, in North Carolina. The NAEMS efforts (Grant et al., 2016) observed an average daily mean emission of 4.4 and 86.1 g NH₃ an⁻¹ d⁻¹ from finisher and sow lagoons in North Carolina, respectively. While the emissions of the NAEMS North Carolina finisher study were about 1.5 times larger than the previous USDA study in 1997-98, the sow-farm NAEMS emissions were about 18.3 times larger than that of the USDA study. The increase in estimated lagoon emissions suggested the swine industry's environmental footprint had increased in spite of improvements in production practices and animal efficiency.

Prior to the NAEMS study, there were discrepancies between various agencies' measured emissions (Aneja et al., 2000; Harper et al., 2004; McColloch, 1999; Todd et al., 2001) on the same farm site. These differences can be partially attributed to measurement technique differences (Harper, 2005). However, the USDA study found that output N from their study farm [including lagoon, housing, and field NH₃ emissions; lagoon chemical conversion of NH₄⁺ to dinitrogen (N₂) gas; field denitrification N₂ and nitrous oxide (N₂O) emissions; crop N use; protein N leaving in animals; and lagoon N storage] accounted for 95% of the incoming feed N, which was taken as a strong validation of their NH₃ emission measurements. While Grant et al. (2016) suggested the large differences between the USDA and NAEMS studies “may be due in part to the measurement approach and meteorological conditions on specific days of measurement”, it is troublesome that the NAEMS emissions (likely to be used for compliance) for finisher and sow farms are significantly larger than other studies in North Carolina. Also, the NAEMS study suggested between 14% to 42% of the finisher and between 50 to 99% of the sow excreted N, respectively, were lost as NH₃. Considering the other forms of N leaving the farm as described above, the NAEMS measured lagoon emission rates suggest that more N may leave the farm than as enters the farm. Based on the USDA findings of chemical conversion of lagoon NH₄⁺-N to N₂-N gas [which may range from 10 to over 40% of input feed N (Harper et al., 2004; Weaver et al., 2012; Harper et al., 2014)], the amount of NH₃ emissions suggested by the NAEMS study seems to be in error. The increase in emissions described in the NAEMS study, compared to the earlier North Carolina studies, is also confusing because there has been a considerable reduction in the N content of feed by the use of synthetic amino acids to replace intact proteins in the diet. Also, improvements have been made in feed efficiency by decreased feed particle size along with animal genetic improvements which has reduced feed required for gain and thereby dry-matter excretion⁵.

⁵ Coffey, M.T. 2018a. Changes in feed utilization and manure output over time. (*unpublished report*)
Coffey, M.T. 2018b. Industry-wide feed efficiency and dry matter excretion. (*unpublished report*)

The use of a limited number of studies to make conclusions about long-term changes in swine NH_3 emissions on a state-wide basis is concerning: these studies used different farms exposed to different environmental conditions, varying measurement durations, different measurement methodologies, and with all farms having different production practices. However, since NH_3 emissions are the result of measurable chemical and environmental factors, it is possible to estimate emissions using knowledge of lagoon chemical characteristics, the region's climatic data, and a validated process model. Ammonia emissions from lagoons have been modeled using statistical regressions (Harper and Sharpe, 1998; Aneja et al., 2000), semi-empirical functions (Grant et al., 2013), and detailed theoretical process schemes in North Carolina (DeVisscher et al., 2002; Bajwa et al., 2006) and Europe (Sommer et al., 2006). Statistical emissions models have little transportability between locations since they depend on the location/time-specific differences of the variables used in the statistical regressions. Models developed from fundamental concepts provide the greatest accuracy across climates and geographical areas. Consequently, modeling of the emissions into the turbulent atmosphere requires a physical understanding of transport processes so the dominant processes can be theoretically described for a wide range of production and environmental conditions. The process (DeVisscher et al., 2002) and statistical (Harper et al., 2004) models used were both validated using emissions from Georgia (Harper et al., 2000) and North Carolina (Harper et al., 2004) finisher and sow farms. The process model accounted for 70% of the variability in daily mean emissions whereas the statistical model accounted for 77% of the variability with inputs of windspeed, lagoon temperature, lagoon ammoniacal N [AN ($\text{AN} = \text{NH}_3 + \text{NH}_4^+$)], and lagoon pH. The statistical (linear) models of Aneja et al. (2008) for summer and winter showed correlation of NH_3 flux (as measured by a chamber technique) with total lagoon N (total Kjeldahl N, TKN).

The North Carolina swine industry has made considerable efforts to enhance production efficiency and reduce its impact on the environment since the USDA and NAEMS studies were conducted. The swine industry has hypothesized that improved feed efficiency, reduced excretion, and improved animal genetics have resulted in a significant reduction of N excretion and NH_3 emission rates over the last 20 years. Since the discrepancies in emission rates between the 2009 NAEMS and earlier studies make it impossible to evaluate whether swine industry improvements have led to a reduced environmental footprint, the purpose of this study was to use emissions models, coupled with archives of lagoon chemical sampling, atmospheric deposition data, and climate data, to evaluate swine emissions since 2001, and to determine the effect of what changes in production practices and efficiency have had on NH_3 emissions and NH_4^+ deposition.

Objectives: Deliverables of this research will include an assessment to evaluate whether or not the swine industry has reduced its NH_3 emissions while at the same time provided a safe and economical food source for the country along with solid local and state economy support.

Materials and Methods:

The swine industry is required by the North Carolina Department of Environmental Quality (NCDEQ) to sample manure lagoons periodically (approximately bi-monthly) for analysis of nutrient content by the North Carolina Department of Agriculture and Consumer Services (NCDA&CS). Included in the nutrient sample analysis are lagoon total N and pH (which affect the physical chemistry of NH_3 in solution and its ability to volatilize from the lagoon). These data are archived by the Office of Land and Waste Management (Anon, 2019a) and presented to the NCDEQ for analysis. Archived data were obtained from 182 lagoons representing 81 finisher and 78 sow primary lagoons along with 5 finisher and 18 sow secondary lagoons in the rural North Carolina Coastal Plain for the period from 2001 to 2018 (representing about 106,000 laboratory analyses for finisher and sow plus primary and secondary lagoons). For this study, the focus was long-term trends in finisher and sow lagoons' NH_3 emissions, and the primary and secondary (overflow) lagoons were analyzed separately.

The selected lagoons were carefully chosen to limit the possibility that outside factors would bias the statistical analysis. For example, any lagoon that changed use (ex. from nursery to finisher operations) during the data analysis period was eliminated. Also, at least 10 years of data needed to be available for any lagoon to be included in the analysis. Any lagoon that had a period of closure was eliminated, unless the exact date of closure was known and more than 10 years of consecutive data were available before the closure. Finally, all known processing and clerical errors were removed, as well as duplicate data entries.

Ammonia emissions ($\text{kg NH}_3\text{-N ha}^{-1} \text{d}^{-1}$) were estimated using a process model (DeVisscher et al., 2002) based on physical and chemical factors affecting volatilization. The model inputs include lagoon AN and pH, windspeed and air temperature from weather data (Anon, 2019b), and water temperature of the emitting surface [which can be estimated from climate data and air/water temperature models (Grant et al., 2016; Weaver et al., 2012)]. While the DeVisscher emissions' model uses AN, the NCDEQ data set only contained total N (TKN) in solution. Grant et al. (2013) proposed using a factor of 90% to calculate AN from TKN in swine lagoons for NH_3 emission calculations, a value close to our findings in this study, which is used in our model calculations. Emissions were also calculated using other semi-empirical or statistical models (Aneja et al., 2008; Harper et al., 2004).

To evaluate the data quality of the samples submitted by the industry, samples were collected by the authors at 12, randomly-selected farm sites taken from the archived farms using a web-based random number generator (Fig. 1A). The lagoons were sampled simultaneously and with the same technique as in the normal sampling process. One lagoon was extensively sampled by the authors to assess both sampling and analytical variability along with spatial variability of the lagoon. Six samples were uniformly collected around the edge of the lagoon and another five samples were uniformly collected in the middle of the lagoon to evaluate spatial variability (Fig. 1B). Additionally, at one of the edge locations, nine samples were consecutively sampled to address both sampling and analytical variability. The samples collected by the

industry were taken by facility personnel and were submitted and analyzed in the normal manner by the NCDA&CS. Samples obtained by the authors were analyzed by a contracted laboratory.

Extreme or erroneous lagoon sample data can be caused by many things such as equipment malfunction, contaminated sample, or a simple clerical error. These outliers can lead to bias in statistical results and ideally should be removed. For this large data set, an iterative approach was developed to identify outlier data-points.

A report (Aneja et al., 1998, Fig. 2) suggested a large increase (about 100%) in NH_4^+ deposition in the rural Coastal Plain of North Carolina during a period largely overlapping the period of swine production expansion. This finding was based on National Atmospheric Deposition Program (NADP) deposition data from the period 1989-1996. For this study, annual, monthly, and weekly NADP data (NADP, 2018) over 40 years (1979-2019) were obtained to examine deposition before, during, and after the period of swine-production expansion (1990-1996). Statistical trend lines of all periods were developed for the NH_4^+ deposition data and variability of input data was evaluated. Trends in the deposition data were examined in relation to both swine (NCDA&CS, 2019) and human (NCOSBM, 2018) population increase.

Results and Discussion:

Lagoon variability: The archived pH and TKN data reveals large variability within the complete data set (105,739 points). To test whether part of the variability might be due to actual spatial variability, 11 samples were taken from various sites within a single lagoon. However, in our sampling evaluation survey (spring, 2019) we found that the N concentrations and pH from the near-shore and the middle of the lagoon sampling sites were not statistically different (Table 1). We thus concluded that one sample is suitable for representing lagoon chemistry for emissions' estimates.

Because variability imparts uncertainty, it is important to characterize all variability components in the industry-collected data to generate valid lagoon-chemistry trendlines. Sources of variability include the following: 1. sampling variability (differences in samples taken at the same place and time), 2. laboratory analysis variability (differences in laboratory results of the same sample), 3. spatial variability (differences in results of samples collected at different places in a given lagoon), 4. season variability (differences in results of samples collected at the same place throughout the year) and 5. variability between lagoons (difference in results between lagoons taken at the same time). Each of these sources affect the ability to determine lagoon chemistry trends. We took nine samples from a single shore site to determine site sample-collection variability and sample-analysis variability in the laboratories. Spatial (including laboratory analysis and sampling) variability was determined by calculating the relative standard deviation (RSD) from both shore and middle-of-the-lagoon sites (Table 2). In order to estimate season variability, sample results from another lagoon (from the industry/NCDEQ data base) were used to calculate yearly RSDs and then averaged. For each year of available data, the standard deviation and RSD were calculated, and those

yearly results were averaged. The seasonal variability (RSD = 28.5%) was found to be much larger than any of the other sources of variability. The RSD values for seasonal variability are 10 to 25 times larger than the RSD values for sampling/analytical variability and 5 to 10 times larger for spatial variability. Thus, the biggest factor within a lagoon is seasonal variability. Such large seasonal variation in the data prevents determination of statistically significant trendlines using bi-monthly data. This seasonal variation is eliminated by using yearly averages that do not suffer from seasonal variation.

Comparing results between the samples collected by the farm personnel and those collected by the authors (Table 3) evaluated the variability and bias between collection systems and with different laboratories. The average of the contract lab TKN values were 80 ppm lower but pH was 0.11 units higher. While a 0.11 pH unit difference appears small, when used to calculate the amount of AN in the NH₃ volatile form, this difference results in 29% more NH₃ in solution. When these two offsetting differences are combined, the net effect was that there was no statistical difference in available NH₄⁺ between the samples collected by farm personnel and the authors. This comparison of results between the sampling and analysis techniques validates the NCEPD data and assures its usefulness in estimating trends in lagoon NH₃ emissions from 2001 until the present.

Statistical Methods: Upon scrutiny of archived data, it became apparent that occasionally some of the values were unrealistically high or low (ex. Fig. 2A) and can be considered improbable given the buffering capacity of lagoons. Extreme or erroneous data can be caused by many things such as equipment malfunction or a simple typographical error or it is possible that many of the outliers came from samples contaminated with solid waste. Changes in lagoon chemical composition occur, but only over an extended period of time, not in sudden spikes. These outliers create bias and may impart uncertainty in many statistical results including means, standard deviations, and trends, and should be removed. Visually identifying extreme data is subjective and impractical (given the quantity of data). Simplistic approaches such as blanket cutoff values or elimination of data that are a specified number of standard deviations away from the mean may reduce subjectivity and are easy computationally, but the mean and seasonal variability are different between lagoons. What may be an outlier in one lagoon may not be in another. Furthermore, lagoons can change gradually over time and using 20 years of data to computationally find outliers is not appropriate since the mean and standard deviation will also change in time. For this large data set, another approach was developed to analyze data on a lagoon-by-lagoon basis using means and standard deviations determined for several seasons at a time rather than using the entire 10 to 20-year span of data. An Iterative Outlier Identification (IOI) method⁶ was developed where extreme outlier classifications were four standard deviations from a five-seasonal mean and these calculations were performed one lagoon at a time. The IOI

⁶ Duffin, S.M., K.H. Weaver, and L.A. Harper. 2019. Analyzing variability of trends and outliers in environmental data. J. Environ. Qual. (*in preparation*)

method also takes into account that when large outliers are present, the calculated standard deviation may be overestimated and a smaller outlier can be missed (Fig. 2A). The process is thus iterative until all extreme outliers are identified (Fig. 2B) and a data trend is obtained without extreme outliers (Fig. 2C). Using the IOI method, 63 of the 20,556 pH and N data pairs were eliminated.

There is normal annual variability of emissions in addition to sampling, analysis, clerical, etc. variability which may be reflected in a long-term trend. A two-year period of data was randomly selected and plotted for both finishers and sows to evaluate annual input factors' variabilities and how they affect NH₃ emissions (Figs. 3A and 4A). Included are the physical and chemical factors most directly related to NH₃ volatilization: lagoon N content (Fig. 3B and 4B), pH (Fig. 3C and 4C), and climate factors of air and lagoon temperature and windspeed (Fig 3D and 4D). Finisher and sow primary lagoon NH₃ emissions (Figs. 3A and 4A) varied similarly on an annual basis. Using all emissions data for comparison with the physical and chemical factors affecting emissions, lagoon pH had the highest statistical correlation, followed by lagoon temperature, and lagoon AN (all $\alpha \leq 0.01$). There was no statistical correlation between windspeed and lagoon NH₃ emissions.

Ammonia Emissions from Manure Lagoons. Ammonia emissions from the ensemble of 182 North Carolina swine farm lagoons were calculated using the USDA process model (DeVisscher et al., 2002). Calculations were based on lagoon chemistry data along with average climate data for each collection site and date. From this information, the average daily emission for the day of each sample collection was calculated (Fig. 5). The dominant pattern in Fig. 5 is the annual trend where emissions are higher in summer than winter. Less visible is the overall downward trend in emissions with time (see polynomial lines, solid for finisher and dash for sow lagoons).

The NAEMS studies in North Carolina (Grant et al., 2016) suggested that NH₃ emissions from the swine industry were considerably larger compared with those found 10 years prior by the USDA (Harper et al., 2004; Harper and Sharpe, 1998) showing emissions from finishers 1.5 and sows 18.3 times larger. However, long-term measurements of lagoon chemistry and modeled NH₃ emissions (since 2001) do not indicate increased NH₃ emissions nor increase (since 2006) in output of other manure chemical components (Table 4), except for finisher primary lagoon Cu. Ammonia emissions decreased for both finisher and sow farm primary and secondary lagoons (Fig. 5A) along with decreases in lagoon N (Fig. 5B) and a slight decrease (more acidic) in pH (Fig. 5C) [Note: For a decrease in 0.1 pH units in solution, there is 26% less NH₃ in solution to volatilize].

While there has been criticism of the swine industry for producing a significant disproportion of total NH₃ emissions on the Coastal Plain of North Carolina, long-term trends of lagoon chemistry composition and resulting NH₃ emissions have not been previously evaluated. The swine industry has improved production efficiency over the years, which has reduced the industry's environmental impact.

Management changes were introduced to reduce excreted nutrients by using synthetic amino acids to improve N and protein efficiency, phytase to decrease phosphorous (P) requirement, reduced feed grain particle size to improve feed digestibility, and swine genetics to improve feed conversion and increase daily weight gain. Figures 7A and 7B show results of management improvement⁵ (since 2009) providing a - 8% improvement in feed conversion to animal weight (Table 4) while producing an average daily animal gain increase of +22%, a reduction in fecal N by 34%, and a reduction in fecal P by 62%.

In contrast to the NAEMS' studies which suggested an increase in NH₃ emissions during the 10 years following a USDA evaluation of North Carolina and Georgia lagoons, there has been a significant reduction in nutrient output as the industry has implemented improved production and management practices along with improved animal genetics during the ensuing years since 2001. The USDA process model (DeVisscher et al., 2002) determined relative NH₃ emissions' decreases (Table 5) for finisher and sow primary lagoons of 47% and 22% and decreases for secondary lagoons of 49% and 50%, respectively. A statistical model by Harper et al. (2004) determined a relative decrease in NH₃ emissions for finisher and sow primary lagoons of 28% and 11% and for secondary lagoons of 19% and 30%, respectively. A statistical model by Aneja et al. (2000) determined much larger relative decreases in in primary lagoon emissions of 54% and 33% for finishers and sows, respectively (the model was not applicable to secondary lagoons). Absolute emissions were different between models, but *all* models using archived historical chemical and climate data showed significant decreases in NH₃ emissions since 2001 ranging from 11% to 54%.

Another factor, which is often ignored, that affects the N content of waste-management systems is thermodynamic conversion of NH₄⁺ to benign dinitrogen gas (N₂), which can affect the annual variability of N content in lagoons. Harper et al. (2004) in a study of two North Carolina farms (farrow to finish and farrow to wean) found N₂ emissions of over 40% of input feed N. In another study of six farms, Harper et al. (2014) in North Carolina showed thermodynamic conversion rates of NH₄⁺ to N₂ gas in different types lagoons (sow, farrow-to-finish, and finisher) ranging from 10% to over 20% of input feed N. Other studies (Harper et al. 2000, 2010; Weaver et al., 2012) have shown significant seasonal amounts of NH₄⁺ converted to benign N₂ in swine lagoons. Little consideration in the literature and by environmental organizations has been given to the reduction of lagoon N by this mechanism as it has been assumed that all the N going into manure treatment lagoons leaves only as NH₃ (Hatfield et al., 1993; Doorn et al., 2002; Grant et al., 2016).

Changes in Lagoon Chemistry: Advancements in swine production practices, changes in feed formulation, improved swine genetics, reduced nutrient excretion⁵, and other management changes have resulted in reduced nutrients in both primary and secondary lagoons (Fig. 6, Table 4) ranging from 17% (N, Sows Primary lagoon) to 95% (Zn, Finishers, Secondary lagoon) except for an increase in Cu in finisher primary lagoons. Copper is an essential dietary element (Holman and Chenier, 2015) and it is known to have

antimicrobial properties (Cromwell, 2001; Cromwell et al., 1998). It is a common industry practice for concentrations of Cu above the minimum nutrient requirement to be fed for growth and feed efficiency enhancement and as an alternative to growth-promoting antibiotics in the growing/finishing phase of production. Copper in primary lagoons of finisher farms increased by 41% since 2006; however, the content in sow primary and both finisher and sow secondary lagoons were not affected and the content percentage decreased.

Climate Impacts: While there was significant correlation between ambient and lagoon temperature with NH₃ emissions ($\alpha = 0.05$), these studies' modeled emissions showed no correlation with windspeed (discussed previously) due to variability. While windspeed is important, it can't be predicted nor used to predict changes in NH₃ emissions. Air temperature has been reported (Robinson, 2019) to have increased about one-degree Fahrenheit (0.6 °C) over the last quarter century in North Carolina⁷. Modeled lagoon water temperature values were increased systematically in relation to a one °C air temperature increase and used with archived lagoon chemical measurements and monthly average windspeeds over the course of the 20-year study. The modeled NH₃ emissions from the hypothetical temperature increase were found to increase about 12% and 8% for finisher and sow lagoons, respectively. Other studies (Grant et al., 2013) predicted if the climate warmed across the country one °C, that finisher and sow farms' emissions would increase NH₃ emissions by 8.0% and 9.2%, respectively. Consequently, climate can have a significant impact on lagoon emissions. It is, however, interesting to note that while a one °C temperature increase in NC would predict an 8-12% increase in emissions, the *actual* NH₃ emissions decreased (ranging from 22% to 54%, depending on model used and in spite of the climate-change temperature increase in North Carolina). This decrease emphasizes the importance of the industry's increased feeding efficiency along with swine genetics and other management improvements that have improved chemical conditions in the lagoons creating lower emissions.

Nitrogen deposition. Ammonia concentrations in the atmosphere, as measured by NADP rainfall deposition (NADP, 2018), were reported (Aneja et al., 1998) to increase in the rural atmosphere of eastern North Carolina between 1989 and 1996. This increase was attributed to swine production expansion from 1990 to 1996. As a result of anticipated detriment to the environment and other concerns, the North Carolina legislature enacted a moratorium on new swine farms in 1997. Initial analysis of the data suggested, using selected time periods (Aneja et al. 1998, Fig. 2; 2003, Fig. 6), an increasing trend in NH₄⁺ deposition by over 100% in the rural area of Sampson County, North Carolina (NC35 near Clinton), during the period of

⁷The air temperature (measured at two meters) linear trend line from the North Carolina Climate Office, Weather and Climate Database, Station KFAY, Climate Division: NC06-Southern Coastal Plain, from 2000 to 2019 shows a 0.66 °C trend increase in air temperature (Anon., 2019b).

swine farm expansion. However, when the entire archived NADP data set for rural, North Carolina, (for 40 years) is taken into account (Fig. 8), the effect of swine expansion on deposition is questionable (as reflected by the statistically significant constant increase in the long-term sites' depositions).

NH₄⁺ Deposition vs. Time: When yearly rural NH₄⁺ deposition is plotted with respect to time using all available data (Fig. 8), there is a strong positive statistical linear trend ($p \ll 0.001$, $R^2 = 0.84$, slope = $0.012 \text{ mg L}^{-1}\text{yr}^{-1}$). Also, a visual inspection of the NADP deposition data (Fig. 9) reveals that the rate-of-increase over the entire available data period does not appear to change significantly regardless of the dynamics of the swine population and the production improvements resulting in reduced NH₃ emissions. When the data prior to the swine expansion (1979-1989), during the expansion (1990-1996), and after the expansion (1997-2017) periods are tested for linear relationship (correlation), the calculated probabilities (p -values) for pre- and during-expansion periods are too large ($p = 0.18$ and $p = 0.11$, respectively; Table 6) to claim a significant linear relationship between deposition and time. Only the post-expansion period has a significant relationship with time ($p = 0.007$). The years 1990-1996 (inclusive) were chosen as the swine expansion years based on the large increase in swine population between years. The swine population numbers in Fig. 8 are for January of each year, so the dramatic swine population increase for 1991 corresponds to rapid growth in 1990. The years designated as the expansion period is very important to clarify because the variability of the deposition data can lead to very different statistical results just by adding or subtracting a few years. For example, when 1991 to 1998 was chosen (Aneja et al., 2003, Fig. 6), moving the end date to 1998 exaggerated the increase of deposition since extending those two years included the two highest deposition values of an 11-year span.

Since more data values would possibly allow for significant correlations, both monthly and weekly data were also evaluated (Fig. 10). Unfortunately, the use of monthly and weekly data did not lead to statistically better linear relationships. In many cases, the p -values for the correlations were smaller with more data, but R^2 values were also smaller, making the variability explained by the linear relationship practically negligible. Results were even less indicative for the urban data where only the total period and post-expansion slopes had significant linear correlation for annual data, and no period had significant correlation for monthly data. Because of variability in the data, a significant change in the rate of deposition during expansion cannot be claimed or disproven. A report by Aneja et. al. (2003, Fig 6) presenting the >100% increase during the years 1991-1998 failed to present rates of deposition increase before, during, and after expansion as well as to test for any statistical differences in these rates. The Aneja et al. report also used data that only partially overlapped the swine expansion period (1989-1996) which cannot be used to reach conclusions about the expansion period of 1990-1996.

Notwithstanding the inability to determine statistical significance of the trends during various time periods, it seems reasonable to expect that the dramatic increase in the swine population would create an increase in deposition, but that production improvements would create a reduction in the deposition rates.

What is noteworthy is that visual inspection (Figs. 8 and 9) shows lack of significant change in deposition after the expansion period. If swine population was the only driving force for the increase in deposition, then a decrease in swine population combined with an increase in swine feed efficiency should have resulted in a reduction of deposition rates in the years post-1996, but this was not observed. The post-expansion rate in deposition is indicative of how non-swine factors affect the rate-increase of NH_4^+ deposition. Consequently, since post-expansion deposition rate did not decrease, then what other factors are affecting this rate of increase in NH_4^+ deposition?

Based on the statistics and observations of the NADP archived data, the major increase in swine population likely contributed in part to an increased rate of NH_4^+ deposition from 1990 through 1996; however, there was a substantial increase in human population also from about 75,000 to about 135,000 persons yr^{-1} beginning in 1990. The continued rise in NH_4^+ deposition after 1996 is likely due to other factors; in particular it is highly probable that human population contributed significantly. It would appear that the suggestions about the environmental effect (Aneja et al., 2000; Walker et al., 2000) of the swine industry as demonstrated by deposition-rate changes, in comparison to human population and other NH_3 emissions' influence, has been overstated.

Summary and Conclusions: Improvement in feed efficiency and management of swine farms have resulted in decreased elemental nutrient output in excretion leading to reduced air-quality emissions. An analysis of archived data collected for the NCDEQ has shown considerable improvement in the swine industry's nutrient output with substantial decreases in nutrient concentrations in manure processing lagoons (with the exception of Cu from finisher animals). All methods of estimating NH_3 emissions based on lagoon nutrient content and climate data, regardless of the models used, show decreased emissions since research began in 1997. Ammonia emissions' models have suggested a decrease from 22% to 54% for finisher and sow, primary and secondary farm lagoons, and depending on which model is used for emissions' calculations. While there is NH_3 emitted by automobiles, industry, municipal waste processing, urban development, and other types of agricultural production, archived NADP data from three time-periods of before, during, and following the period of swine production expansion cannot be statistically evaluated for differences in slopes because of data variability; however, the long-term trends are statistically significant. Ammonium depositions generally had a long-term linear increase in deposition similar to human population increase. Also, there was no statistically significant decline in the increase in deposition in the swine post-expansion (moratorium) period. Post-expansion deposition continued to increase even though swine numbers decreased and the industry made significant improvements in NH_3 emissions, suggesting the increasing deposition was due to NH_3 emissions from sources other than swine. Analysis of the long-term NADP deposition data indicated that a significant portion of the increase in rural and urban NH_4^+ deposition over

the last 40 years, which had been attributed to swine NH₃ emissions, is likely correlated with human population increase and its associated NH₃ emissions, particularly after the swine moratorium.

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Table 1. Differences in chemical composition between lagoon sampling locations (shore vs. lagoon middle). There was no significant difference between shore and middle data.					
	Average shore ± StDev (n=6)	Average middle ± StDev (n=5)	Absolute Difference	Percent Difference	Significant ($\alpha = 0.05$)
pH (pH units)	7.71 ± 0.05	7.74 ± 0.02	0.03	0.4%	No
TKN (ppm)	452 ± 27	431 ± 11	20	3.2%	No

Table 2. Use of relative standard deviation (RSD) to characterize variability in chemical data.		
Parameter	pH (pH units)	TKN (ppm)
Sampling/Analytical Average	7.70	469
Sampling/Analytical Standard Deviation	0.02	5.4
Sampling/Analytical RSD	0.3%	1.1%
Shore Spatial Average	7.74	445
Shore Spatial Standard Deviation	0.05	30
Shore Spatial/Analytical RSD	0.65%	6.7%
Middle of Lagoon Spatial Average	7.71	431
Middle of Lagoon Spatial Standard Deviation	0.02	14
Middle of Lagoon Spatial/Analytical RSD	0.3%	3.2%
Single Lagoon Study Average	7.56	467
Single Farm Standard Deviation	0.23	128
Single Farm RSD	3.0%	28.5%

Table 3. Difference in analysis results between employee- and researcher-collected samples.				
Parameter	Average difference between industry and research samples (n=12)	Relative difference between industry and research samples (n=12)	Level of Significance (α)	Significance
pH (pH units)	0.11	1.5%	0.05	Yes
TKN (ppm)	-80	-15%	0.001	Yes
[NH ₃ (aq)] (ppm)	1.9	13%	0.05	No

Table 4. Changes in the swine nutrient content and NH₃ emissions ($p < 0.05$ except where marked) of waste processing lagoons due to improved production practices over the period from 2001 up to 2019. Annual averages were used when data exhibited seasonal variation.

Animal and Lagoons	NH ₃ [†] (kg NH ₃ ha ⁻¹ d ⁻¹)	N [†] (ppm)	pH [†] (pH units)	P [‡] (ppm)	K [‡] (ppm)	Cu [‡] (ppm)	Zn [‡] (ppm)	Feed Conversion [§] (kg feed kg gain ⁻¹)	Daily Gain [§] (kg d ⁻¹)	Fecal N [¶] (kg pig ⁻¹)	Fecal P [¶] (kg pig ⁻¹)
Finishers Primary	-47%	-35%	-0.10 units	-44%	-34%	+41 ^{**}	-78%	-8%	22%	-34%	-62%
Finishers Secondary	-49% ^{§§}	-27% ^{§§}	-0.20 units	-75%	-62%	-38%	-95%	N/A ^{††}	N/A	N/A	N/A
Sows Primary	-22%	-17%	-0.02 units	-45%	-19% ^{§§}	-65%	-68%	N/A	N/A	N/A	N/A
Sows Secondary	-50%	-56%	-0.02 units	-44%	-32%	-78%	-95%	N/A	N/A	N/A	N/A

[†] Available data beginning Feb 1, 2001 [based on calculated NH₃ emissions by a USDA process model (DeVisscher et al., 2002)].

[‡] First data available Feb 3, 2006.

[§] First data available May 1, 2009.

[¶] First data available Jul 6, 2009⁵.

^{††} Not available.

^{**} $p < 0.2$.

^{§§} $p < 0.1$.

Table 5. Changes in NH₃ emissions from 2001 up to 2019 ($p < 0.01$ except where marked) calculated by a process model (DeVisscher et al., 2002) and empirical models (Aneja et al., 2000; Harper et al., 2004) using archival lagoon data (Anon, 2019a).

	DeVisscher et al., 2002	Harper et al., 2004	Aneja et al., 2000
Finishers Primary Lagoons	-47%	-28%	-54%
Finishers Secondary Lagoons	-49% [†]	-19% [†]	N/A [‡]
Sows Primary Lagoons	-22%	-11% [§]	-33%
Sows Secondary Lagoons	-50%	-30% [§]	N/A

[†] $p < 0.1$.

[‡] Not applicable to secondary lagoons.

[§] $p < 0.05$.

Table 6. Swine expansion plus rural and urban NH ₄ ⁺ deposition statistics.				
Period change	<i>p</i> -value	R ²	Trendline	Significance <i>α</i>
Swine expansion				
1979-1989	0.23	NS [†]		No significant linear relationship
1990-1996	0.000005	0.989	$y = 1132143x - 2250575000$	0.001
1997-2017	0.0007	0.464	$y = -46883x + 103451558$	0.001
Rural Deposition				
1979-1989	0.18	NS		No significant linear relationship
1990-1996	0.11	NS		No significant linear relationship
1997-2017	0.007	0.328	$y = 0.007x - 13.82$	0.01
1979-2017	3×10^{-16}	0.839	$y = 0.021x - 23.77$	0.001
Urban Deposition				
1979-1989	0.36	NS		No significant linear relationship
1990-1996	0.67	NS		No significant linear relationship
1997-2017	0.014	0.277	$y = 0.005x - 9.58$	0.05
1979-2017	0.000002	0.466	$y = 0.004x - 8.02$	0.001

[†]Not statistically significant.



Fig. 1. A. Randomly-sampled farms in the Coastal Plain of North Carolina. B. A randomly-selected farm for determination of variability in sampling.

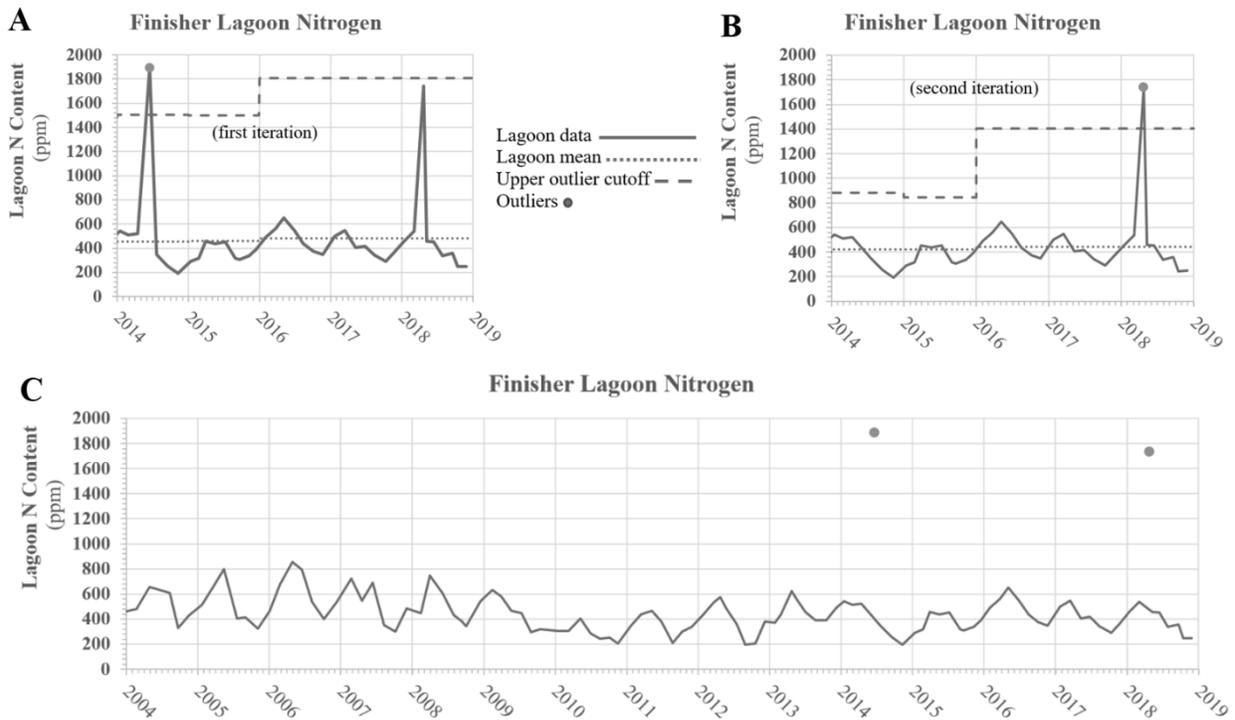


Fig. 2. The Iterative Outlier Identification method used a five-season mean (A) and considered an outlier classification of four standard deviations from the mean thus rejecting one data point. A second iteration (B) eliminated a second outlier producing a normal seasonal data trend (C) with no obvious outliers.

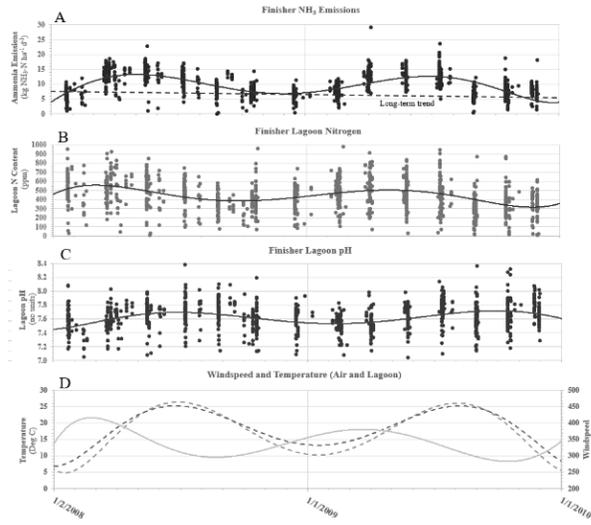


Fig. 3. Variability (finisher production, primary lagoon) of potential effects on (A) NH₃ emissions, (B) lagoon N content, (C) lagoon pH, and (D) climate variation (black dash, lagoon; gray dash, air; line, windspeed).

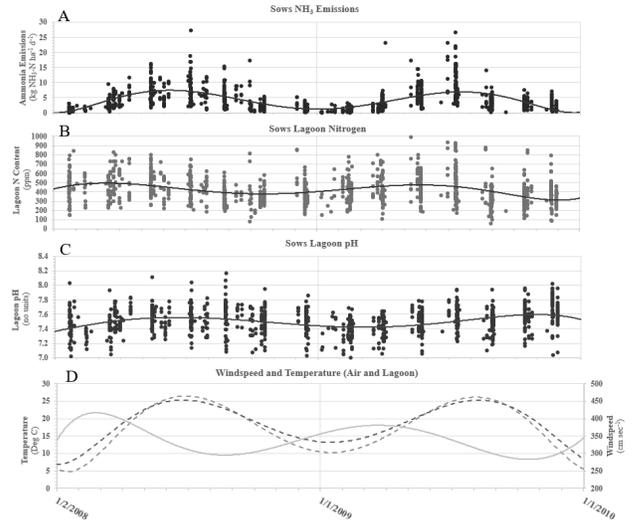


Fig. 4. Variability (sow production, primary lagoon) of potential effects on (A) NH₃ emissions, (B) lagoon N content, (C) lagoon pH, and (D) climate variation (black dash, lagoon; gray dash, air; line, windspeed).

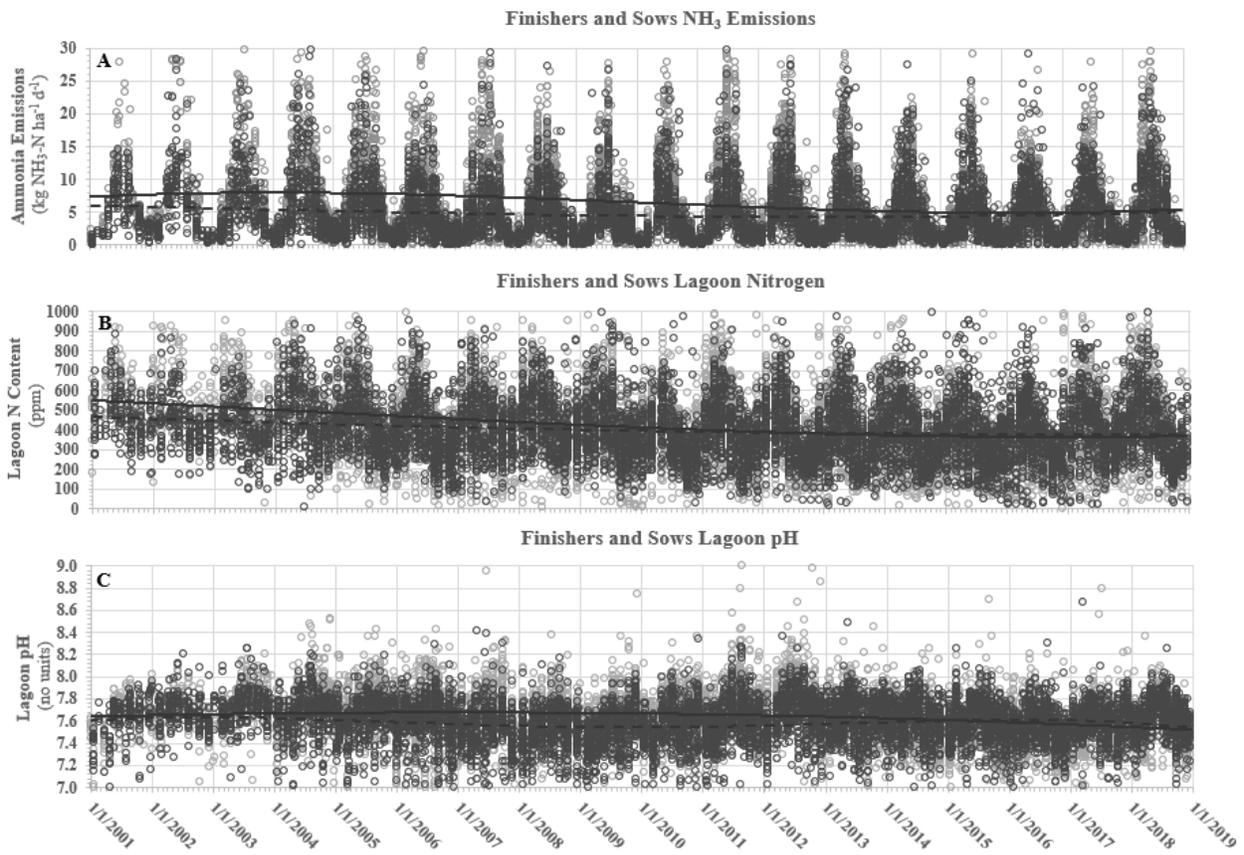


Fig. 5. Emissions from finisher and sow lagoons with chemical factors influencing volatilization, lagoon N content and pH. (Lighter circles and solid lines denote finisher data.)

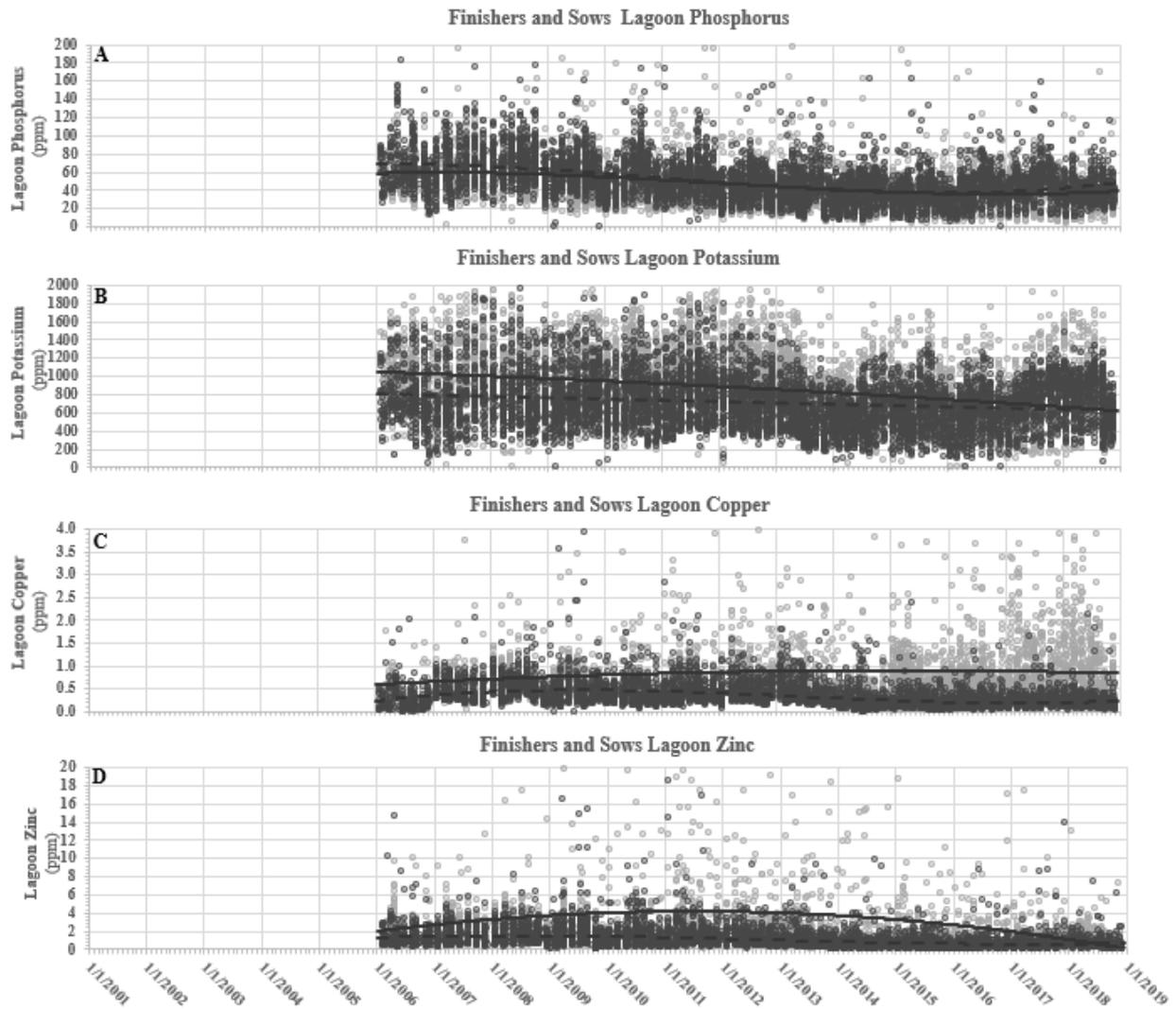


Fig. 6. Decreases in lagoon nutrient content due to changes in swine production (except for finisher Cu content). (Lighter circles and solid lines denote Finisher data.)

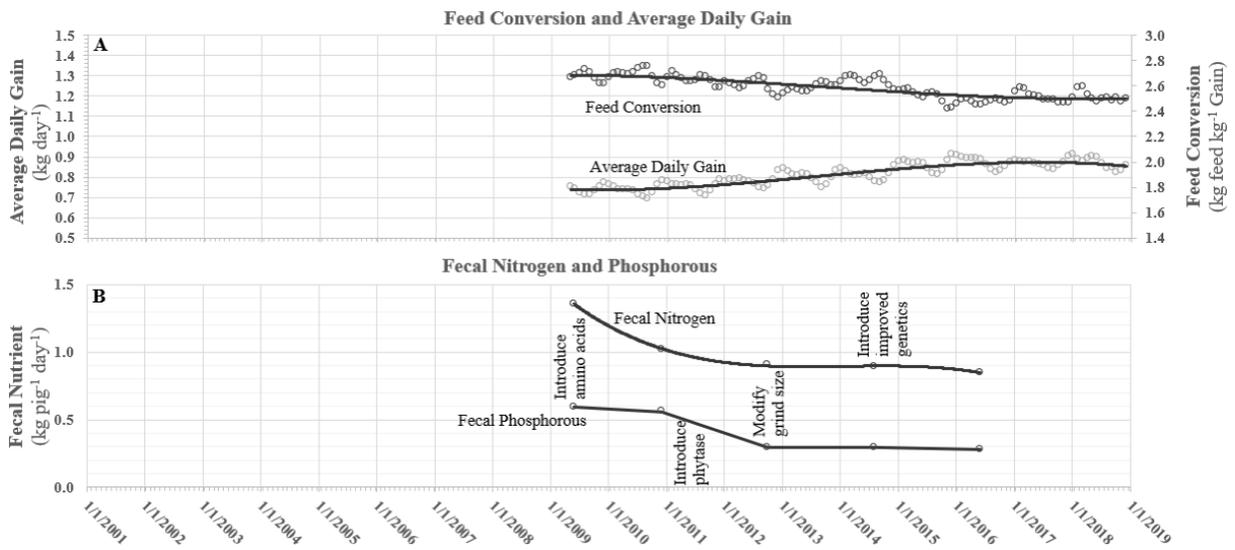


Fig. 7. (A) Changes in feed conversion resulting in increasing average swine daily gain (after Coffey, 2018A⁵) and (B) Changes made in swine production management resulting in fecal nutrient reduction (after Coffey, 2018B⁵)

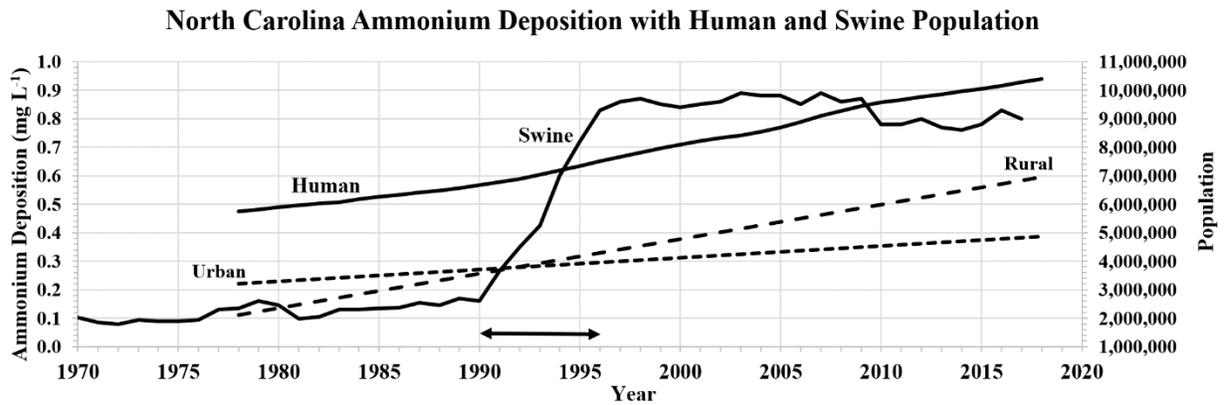


Fig. 8. Rural and urban sites' NH₄⁺ deposition trend-lines along with human and swine population. Statistics of the trend-lines for rural and urban deposition may be found in Table 6 (Arrow denotes swine production expansion period.).

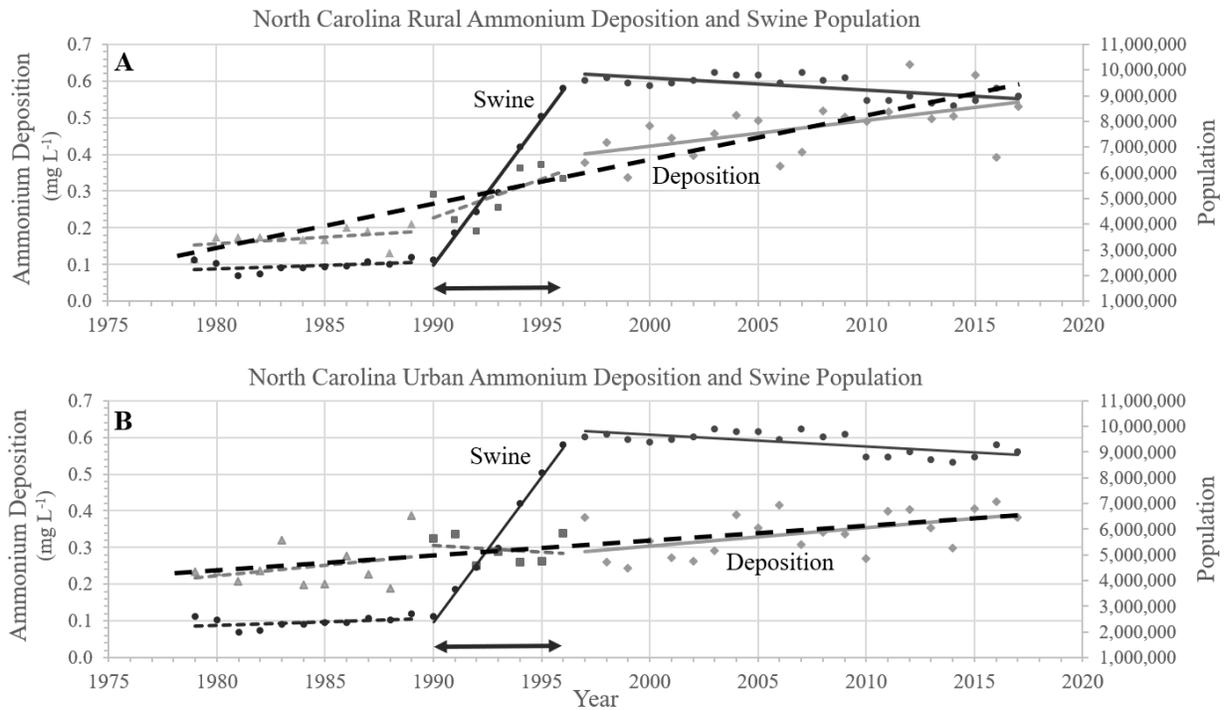


Fig. 9. North Carolina swine population along with NADP ammonium deposition in both (A) rural and (B) urban sites. Solid lines and the long-term deposition line (long dashes) are statistically significant (Table 6) whereas short-dash lines are not statistically significant but are trend (best fit least-squares) lines provided for illustration. Arrows denote the swine expansion period.

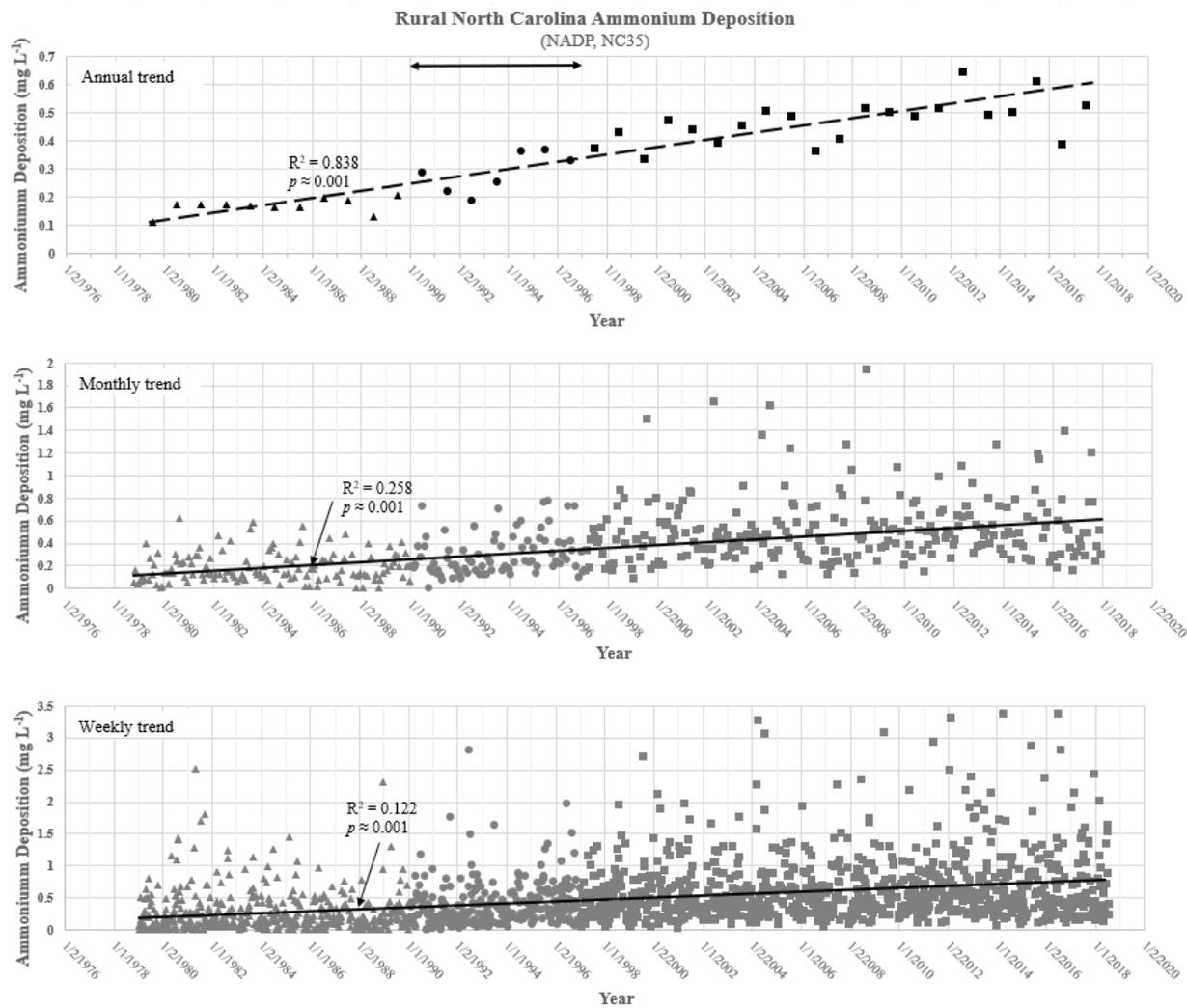


Fig. 10. Annual, monthly, and weekly NADP NH_4^+ deposition trends. Data points are for pre- (triangles), during- (circles), and post-expansion (squares) deposition periods.