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Key Points:
- The National Air Quality Forecast System model overpredicted ammonia at a continuous monitor in a region of intensive animal production
- Model predictions for ammonia were improved by using State permit information for locations of animal feeding operations
- The model underpredicted wet deposition of ammonium ion and dry deposition of ammonia in eastern North Carolina

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(continued)

Evaluating Ammonia (NH_3) Predictions in the NOAA NAQFC for Eastern North Carolina Using Ground Level and Satellite Measurements

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Abstract Ammonia (NH_3) in the atmosphere contributes to the formation of airborne fine particulate matter ($PM_{2.5}$), which is associated with adverse human health effects. The emission, transport, reactions, and deposition of NH_3 in the atmosphere are modeled using the Community Multiscale Air Quality (CMAQ) model, within the U.S. National Air Quality Forecast Capability (NAQFC). The purpose of this current work is to evaluate the capability of the NAQFC CMAQ model and to identify potential improvements to NH_3 emissions estimates and prediction methods. This study focuses on CMAQ predictions of atmospheric NH_3 in North Carolina, including a region with intensive animal production and enhanced NH_3 emissions. The CMAQ model is run for July 2011 using a version of the 2011 National Emissions Inventory in which agricultural NH_3 emissions were adjusted to reflect the lower end of the range of estimates from the current process-based emissions model. The NAQFC CMAQ model overpredicted atmospheric NH_3 at a continuous monitor in Clinton, NC, within the region of intensive animal production. The average concentration measured by the monitor was 6.6 ppbv, while the average predicted by the model was 10.5 ppbv, a 60% overprediction. Outside of the region of intensive animal production, both measured and modeled NH_3 concentrations were low, 1.3 ppbv or less. The model underpredicted wet deposition of NH_4^+ and dry deposition of NH_3. It is believed that the overestimation of NH_3 at Clinton is attributable at least in part to the underestimation of wet and dry deposition in North Carolina.

1. Introduction

Ammonia (NH_3) in the atmosphere contributes to the formation of airborne fine particulate matter ($PM_{2.5}$), which is associated with a number of adverse human health effects, including aggravated asthma, irregular heartbeat, and premature death (Pope et al., 2009). Ammonium compounds, including ammonium sulfates (NH_4HSO_4 and $(NH_4)_2SO_4$) and ammonium nitrate (NH_4NO_3), make up a large fraction of fine particulate matter ($PM_{2.5}$; Kwok et al., 2013). Ammonia can be important in the nucleation of new particles (Holmes, 2007; Herb et al., 2011). These ammoniated particles scatter light, attenuating visibility, and can result in some atmospheric cooling (Pinder et al., 2013). Ammonia also contributes to a cascade of other environmental impacts. Ammonia and particulate ammonium compounds (NH_4^+) in the atmosphere are deposited through both wet and dry processes to terrestrial and aquatic ecosystems, leading to increased levels of biologically available nitrogen, termed reactive nitrogen, in these ecosystems. This can lead to eutrophication of aquatic ecosystems and losses of species diversity (Battye et al., 2017; Jones et al., 2013; Paerl, 1988; U.S. EPA SAB, 2007). A portion of the NH_3 and NH_4^+ is also converted to gaseous nitrous oxide (N_2O) by soil and aquatic microorganisms. This N_2O reenters the atmosphere and absorbs infrared radiation, with a climate change potential approximately 250 times that of CO_2 (IPCC, 2014). N_2O also contributes to stratospheric ozone depletion (Revell et al., 2015).

The National Oceanic and Atmospheric Administration (NOAA) models the emission, transport, reaction, and deposition of NH_3 and its reaction products within the framework of the U.S. National Air Quality Forecast Capability (NAQFC). The NAQFC uses the Community Multiscale Air Quality (CMAQ) model (Byun & Schere, 2006), with emission inputs from the U.S. Environmental Protection Agency (U.S. EPA),

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Environment and Climate Change Canada, and Mexico (Pan et al., 2014; Tong et al., 2015). The bulk of NH_3 emissions to the atmosphere in the United States emanates from agricultural operations, primarily animal waste management and synthetic nitrogen fertilizer application (Aneja et al., 2008; Aneja et al., 2009). The magnitude and distribution of agricultural NH_3 emissions are subject to considerable uncertainty (Battye et al., 2003). Emissions are calculated using emission models coupled with animal population data from the agricultural census (McQuilling & Adams, 2015). Emissions are dependent on waste handling and fertilizer application techniques and on voluntary measures implemented to prevent losses of surplus reactive nitrogen to the environment. Limited information is available on the implementation of such measures. Emission-generating activities are also allotted to different times of the year and to geographic modeling grids using temporal and spatial allocation factors, which add to the uncertainty of model emissions estimates (U.S. EPA, 2015).

Methods are needed to evaluate the capability of CMAQ to predict NH_3 within the NAQFC and to identify potential improvements to NH_3 emissions estimates and prediction methods. Such improvements would also help to improve predictions of reactive nitrogen deposition and $PM_{2.5}$ concentrations in the atmosphere. Some previous validation studies of NH_3 predictions in CMAQ have used secondary indicators such as wet deposition of NH_4^+ ions and the concentration of NH_4^+ in $PM_{2.5}$ (Gilliland et al., 2006; Kelly et al., 2014). The Ammonia Monitoring Network (AMoN) has recently become a source of data on long-term NH_3 gas concentrations across the United States (NADP, 2018) which can be used to evaluate CMAQ NH_3 predictions. However, the density of AMoN sites is limited, with only 55 sites nationwide in 2011, being expanded to about 100 sites in 2015. In addition, the network measures 2-week average NH_3 concentrations and, therefore, does not provide information on short-term variations. Recent satellite infrared spectrometry measurements offer the opportunity to provide a promising source of atmospheric NH_3 concentration data. Satellites can provide spatial coverage and resolution superior to the ground-based measurement network (Whitburn et al., 2016).

This current study evaluates CMAQ predictions of atmospheric NH_3 in North Carolina, with a particular focus on Eastern North Carolina. This is a region of high NH_3 emissions due to a concentration of Confined Animal Feeding Operations (CAFOs) for swine and poultry production. In fact, Sampson and Duplin counties in eastern North Carolina were estimated to have the highest rates of atmospheric NH_3 emissions (per land area) of all U.S. counties in the 2011 and 2014 National Emissions Inventory (NEI; U. S. EPA, 2015, 2018b). The North Carolina Department of Environmental Quality (NCDEQ) made hourly measurements of atmospheric NH_3 at three sites in North Carolina (Shandrikar et al., 2006). The longest operating site, at the Clinton Crop Research Center in Sampson County, operated from 2004 to 2015. The NCDEQ also maintains a permit database showing the locations of animal waste treatment lagoons (NCDEQ, 2017). This information provides a unique opportunity to evaluate model performance toward potential improvement of spatial allocation used in the NEI for NH_3 emissions.

In the current study, the NAQFC CMAQ model is implemented for the month of July 2011 using a modified version of the 2011 NEI. Although confined to a rather short study period constrained by data availability, we conducted a meteorological study for the major state variables over the Clinton monitor and confirmed that the month of July was representative of the summer condition there for 2011. Predictions are evaluated against several sets of atmospheric measurements: hourly concentrations of atmospheric NH_3 from the Clinton monitor, biweekly average atmospheric NH_3 concentrations measured at three AMoN sites in North Carolina, atmospheric concentrations of NH_4^+ in particulate matter at monitors collocated with the AMoN sites, and total atmospheric column loadings of NH_3 retrieved from measurements by the Infrared Atmospheric Sounding Interferometer (IASI) on the Metop satellite (Van Damme et al., 2017; Whitburn et al., 2016). Predictions of NH_4^+ deposition from the second (adjusted) model run are also compared with measured deposition at eight sites. Figure 1 shows the locations of the different monitoring sites used in this case study. The figure also highlights the location of Sampson and Duplin counties, which have the densest concentration of NH_3 emissions as in the 2011 NEI.

This study is the first use of hourly NH_3 concentrations from the Clinton monitor in Sampson County to evaluate predictions by the CMAQ model. The Clinton Crop Research Center is located within a region of dense animal population but does not itself include a CAFO. This evaluation provides information on the performance of the model for reproducing diurnal patterns of NH_3 concentration. In addition, this study

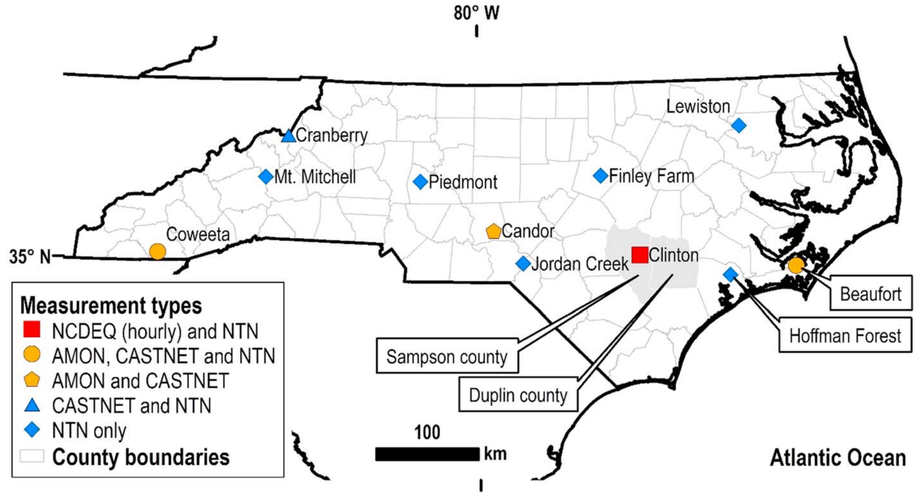

Figure 1. North Carolina case study domain, showing locations of measurement sites. NCDEQ = North Carolina Department of Environmental Quality; NTN = National Trends Network; AMON = Ammonia Monitoring Network; CASTNET = Clean Air Status and Trends Network.

uses improved spatial information which is available for NH_3 emission sources in North Carolina and many other states.

2. Data and Methods

2.1. Air Quality Model

CMAQ model version 5.1 (Appel et al., 2017; Byun & Schere, 2006) was used to predict air pollutant concentrations and deposition for the continental United States in July 2011. The configuration of the CMAQ model within the NAQFC is described in more detail in Tang et al. (2017, 2015). Meteorological predictions to drive the air quality model were generated using the Weather Research and Forecasting Advance Research WRF regional meteorological model, version 3.4.1, with the ACM2 planetary boundary layer scheme. The horizontal resolution of both models is 12 km, with 42 vertical layers with a domain top at 50 hPa (about 20.5 km). The height of the lowest vertical layer was 8 m above the ground. The gaseous chemistry is based on Carbon Bond 2005 e51 chemical mechanism (Appel et al., 2017), and aerosol chemistry is based on the AERO6 module of CMAQ version 5.1. Its dry deposition computed for NH_3 is based on the M3Dry module (Mathur et al., 2005).

The U.S. EPA has developed a bidirectional surface exchange model for NH_3 to be used with CMAQ (Bash et al., 2013; Cooter et al., 2012; Pleim et al., 2013). This model allows for the potential volatilization of NH_3 to the air from vegetated landscapes, offsetting the NH_3 deposition flux and resulting in higher atmospheric concentrations of NH_3. In testing of the bidirectional flux model, predicted atmospheric NH_3 concentrations were 10% higher, on average, than previous predictions with the unidirectional deposition flux approach (Bash et al., 2013; Cooter et al., 2012). This difference is larger in areas with denser NH_3 emissions. NOAA is in the process of incorporating the bidirectional model into the NAQFC version of CMAQ; however, this process has not been completed for the current research.

2.2. Emissions inventory

The NAQFC CMAQ model uses air pollutant emissions from NEI Emissions Modeling Platforms, which are produced by the U.S. EPA every 3 years. The current study is based on the 2011 NEI modeling platform (U.S. EPA, 2016) with some modifications discussed in this section.

Based on the 2011 NEI platform, agricultural operations accounted for 96% of NH_3 emissions in North Carolina, with 88% from emanating from animal waste management operations and 8% from the application of synthetic nitrogen fertilizers. In Sampson and Duplin counties, agricultural sources accounted for over 99% of NH_3 emissions, with 98% from animal wastes and less than 2% from synthetic fertilizers (U.S.

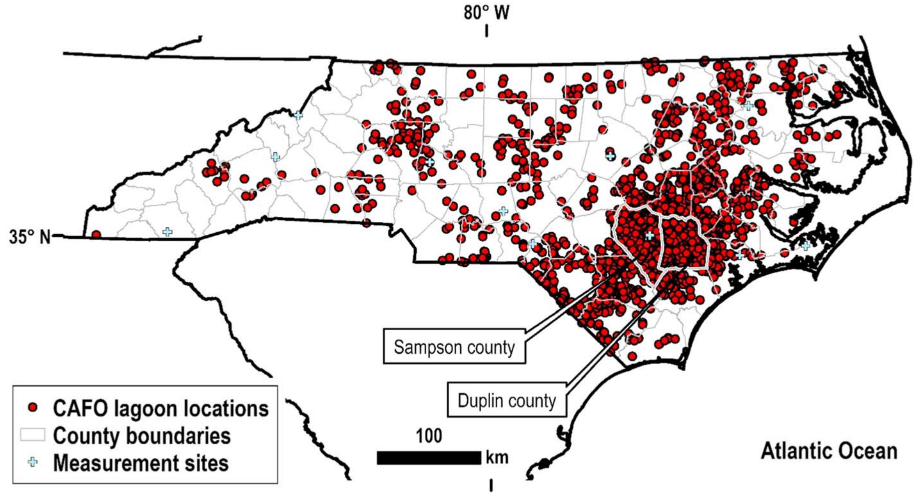

Figure 2. Location of CAFO waste handling lagoons based on state permits. CAFO = Confined Animal Feeding Operation.

EPA, 2015). Swine operations accounted for 73% of the NH_3 emissions in these counties, and poultry operations accounted for 25%.

Within the modeling platform, a standardized national methodology is used to develop the NH_3 emissions inventory for most states, including North Carolina. The methodology has evolved with each iteration of the NEI but always begins with county level animal populations from the Census of Agriculture and Annual Surveys by the Department of Agriculture (USDA, 2019). EPA also derived county level fertilizer consumption statistics from trade association data (U.S. EPA, 2015).

EPA multiplies the animal populations and fertilizer consumption figures by emission factors from the Carnegie Mellon University (CMU) NH_3 model in order to estimate county level emissions (Davidson et al., 2003; U.S. EPA, 2015). The CMU model incorporates emissions from the animal houses and all phases of animal waste handling, including transfer, storage, processing, and application to fields. All of these components are incorporated into a single emission factor for each type of animal, which is temperature dependent, and calculated on a day-specific basis. CMU emission factors for fertilizer do not take temperature into account. Instead, emissions are calculated for an average day of each month based on the typical monthly pattern of fertilizer application. The daily emissions estimates for both animals and fertilizer are allocated to hourly values using hourly temporal allocation factors (U.S. EPA, 2015). County level emissions are allocated to 12-km modeling grids using spatial allocation factors based on the distribution of farm land.

In preliminary testing of the NAQFC CMAQ model with the standard 2011 NEI, NH_3 and particulate NH_4^+ were overpredicted for North Carolina. Therefore, the current study used a lower end estimate for emissions from swine and poultry feeding operations in North Carolina. This lower end emissions estimate was developed based on published error ranges for the NH_3 emissions model used in the NEI (McQuilling & Adams, 2015). The normalized mean error of the model is reported at 28% for swine housing and 61% for waste handling. This yields a weighted average normalized mean error of 55% for swine. Normalized mean errors for poultry range from 28% for broilers to 55% for laying hens. For the current study, NH_3 emissions from swine and poultry operations in North Carolina were lowered by 55% and 40%, respectively, resulting in a 42% reduction in overall statewide NH_3 emissions.

Information compiled by the NCDEQ on the location of CAFOs was also used to improve on the spatial allocation of NH_3 emissions for the current study. We retained the county level distribution of emissions, which are based on animal census data. Within each county, NH_3 emissions were reallocated to 12-km modeling grids using the locations and handling capacities of animal waste lagoons in the county. Information for these calculations was obtained from the NCDEQ CAFO permit list (Figure 2; NCDEQ, 2017). The impact of the change in spatial allocation is particularly significant in the neighborhood of the Clinton measurement site. Based on the distribution of CAFOs in Sampson County, the revised spatial allocation method resulted in a 41% reduction in NH_3 emission density for the CMAQ grid containing the Clinton monitor.

2.3. Ground Measurements

Hourly atmospheric NH_3 concentrations were obtained from a continuous monitoring system operated by the NCDEQ at the Clinton Crop Research Station (NCDEQ, 2016; Shandrikar et al., 2006) between 2004 and 2015. The NCDEQ monitor used a Thermo Scientific Model 17i Ammonia Analyzer. The detection limit of the Thermo Scientific instrument is 1 ppbv, and the precision is 0.4 ppbv (Thermo, 2014). The NCDEQ monitors underwent automated calibration on an average frequency of about once every 36 hr. Following the calibration process, concentrations were frequently unstable for about 4 to 5 hr. Therefore, the NCDEQ NH_3 data were filtered so that results were not used for the first 5 hr after calibration.

Longer-term average atmospheric NH_3 concentrations were also obtained from three AMoN sites in North Carolina. AMoN monitors use passive diffusion collectors, changed every 2 weeks (NADP, 2014). The detection limit of the AMoN passive sampler is approximately 100 pptv for samples collected over a 2-week period, with an accuracy of $\pm$6% (*Sigma* Aldrich, 2019). The AMoN sites are collocated with Clean Air Status and Trends Network (CASTNET) sites which analyze the concentration of NH_4^+ and other ionic species in airborne particulate matter with a precision of $\pm$20% (AMEC, 2013; U.S. EPA, 2018a).

2.4. Satellite Measurements

We compare CMAQ predictions with daytime retrievals of total atmospheric column NH_3 which have been published for the IASI instrument on the Metop-A satellite (Whitburn et al., 2016). The satellite makes daily passes over the region and uses a cross-track scanning system to collect infrared spectra for numerous pixels on each overpass. The footprint of a measurement ranges from 12 km by 12 km at nadir up to 20 by 39 km at the edge of the swath. Ammonia column loadings are retrieved from the spectra using a neural network algorithm which also computes the relative error for each measurement (Van Damme et al., 2017; Van Damme et al., 2018; Whitburn et al., 2016). This IASI retrieval algorithm differs from retrieval algorithms for other satellite systems in that it does not require an a priori assumption for the NH_3 loading.

The current CMAQ comparison uses NH_3 columns calculated from IASI measurements using the ANNI-NH_3-Version 2.2 retrieval algorithm (Van Damme et al., 2017). We interpolated the CMAQ prediction of NH_3 column loading at the location of each available IASI measurement and at the time that the measurement was made. The timing of the IASI measurements in North Carolina during the July 2011 study period ranged from 8:40 to 11:00 A.M. local standard time (UTC−5).

2.5. Deposition Measurements

The National Trends Network (NTN) for precipitation chemistry includes eight sites that were operating in North Carolina in 2011. These sites measure rainfall amounts and concentrations of ionic species, including NH_4^+. The detection limit for NH_4^+ is 0.006 mg/L, and the average difference between replicate measurements of NH_4^+ was 2% for the concentration range measured at the North Carolina sites (Dombek, 2012). CMAQ predictions of wet NH_4^+ deposition are compared with NTN measurements for the July 2011 timeframe.

Dry deposition measurements for NH_3 gas are limited. Phillips et al. (2004) measured the vertical gradient of NH_3 in order to determine dry deposition fluxes in July 2002 at the Finley Farm site in central North Carolina, downwind of a research hog CAFO. Finley Farm is an agricultural research station located at the outskirts of Raleigh, NC, surrounded by a mixture of forest, residential, and low density commercial development. On the scale of the 12-km CMAQ grid, there was no change in the land use pattern in the immediate vicinity of the farm. Temperatures in July 2011 and July 2002 were similar, with an average high temperature of 33 °C in both years and an average low temperature of 21 °C in both years. In 2002, rain was observed on 10 days, while 7 days of rain occurred in 2011 (NOAA, 2019). We compare these with CMAQ predictions of dry deposition with measurements made at Finley Farm.

Table 1
North Carolina Department of Environmental Quality and Ammonia Monitoring Network Measurements of NH_3 Gas Compared With Model Predictions

Monitor site and location	Time frame	Measurement (ppbv)[a]	Model with modified emissions inventory		
			Prediction (ppb)[a]	Absolute bias (ppbv)	Normalized bias (%)
Clinton Research Station (NC95), 35.0258°N, 78.2783°W	Day	9.0 ± 6.6	15.7 ± 8.9	6.7	74
	Night	4.1 ± 2.7	5.3 ± 5.5	1.2	27
	Average	6.6 ± 5.8	10.5 ± 9.2	3.9	60
Beaufort AMoN site (NC06), 34.8846°N, 76.6207°W	5–19 July and 19 July to 5 August	1.3 ± 0.2	0.2 ± 0.3	−1.1	−81
Candor AMoN site (NC26), 35.2632°N, 79.8365°W	5–19 July and 19 July to 5 August	0.8 ± 0.9	1.2 ± 1.0	0.4	54
Coweeta AMoN site (NC25), 35.0605°N, 83.4305°W	5–19 July and 19 July to 5 August	0.4 ± 0.1	0.3 ± 0.6	−0.1	−35

[a]Average with standard deviation.

2.6. Model to Measurement comparisons

Prediction accuracy for the NAQFC CMAQ model was quantified by computing the normalized mean bias (NMB)

$$NMB = \frac{\sum_{i=1}^{N}[C_{\mathrm{mod}}(i) - C_{\mathrm{obs}}(i)]}{\sum_{i=1}^{N} C_{\mathrm{obs}}(i)},$$

where NMB is the normalized mean bias, $C_{\mathrm{mod}}(i)$ and $C_{\mathrm{obs}}(i)$ are, respectively, the model prediction and the observed concentration at a given location and time, and N is the number of observations. Normalized mean bias is commonly used to assess the performance of air quality models (Boylan & Russell, 2006). This parameter focuses on systematic biases in the model, rather than smaller-scale statistical variations.

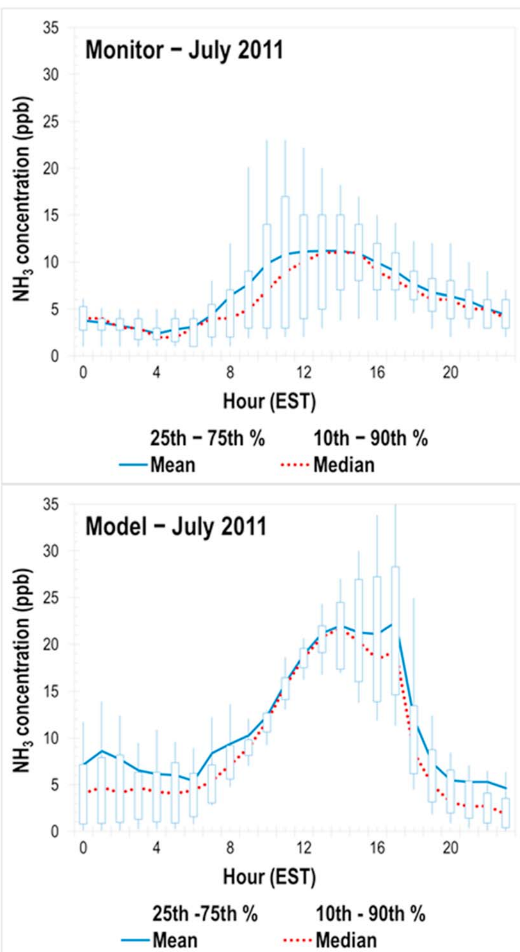

Figure 3. Measured diurnal pattern at the Clinton monitor site compared with model predictions. Solid line and dotted line show the mean and median, respectively. Vertical box shows 25th and 75th percentiles, and vertical lines show 10th and 90th percentiles.

3. Results

3.1. Model Predictions Compared With Ground Level Air Pollution Measurements

Table 1 compares predicted ground level-atmospheric NH_3 concentrations from CMAQ with measured concentrations at the continuous monitoring site in Clinton and at the three North Carolina AMoN sites. The model overpredicted atmospheric NH_3 at the Clinton site by an average of 6.7 ppbv or an NMB of 74% during the daytime. The overprediction was much less at night, an average difference of 1.2 ppbv or 27%. The average overprediction for a 24-hr period was 3.9 ppbv or 60%. Figure 3 shows that the diurnal profile of atmospheric NH_3 predicted by the model is similar to the diurnal profile measured at the Clinton site during July 2011. Both the measured and modeled concentrations increase substantially during the day. However, the modeled concentration shows a secondary peak between 17:00 and 18:00 local standard time. We believe that this peak results from the diurnal emission pattern used in the NEI, which is not based on grid-specific data but on national average temperature patterns. In the NEI temporal allocation, emissions peak at about 13:00 and then taper off gradually until about 20:00. In Eastern North Carolina under the current modeling study, the mixed layer descends prior to this end of the afternoon peak in emissions, causing modeled emissions to be artificially concentrated in a smaller volume. This issue may be alleviated in more recent versions of the NEI, which use local meteorological data for the temporal allocation of NH_3 emissions (U.S. EPA, 2018c).

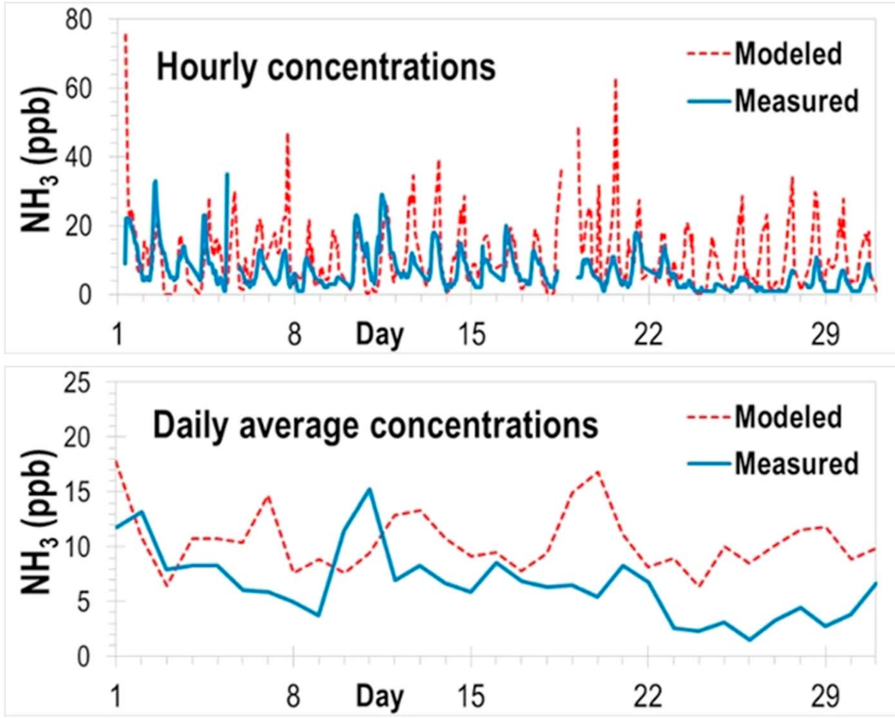

Figure 4. Temporal variations in predicted (with revised inventory) and measured NH_3 concentrations at the Clinton monitoring site.

Figure 4 shows temporal variations in predicted and measured NH_3 concentrations at the Clinton site on an hourly time scale and on a daily time scale during the July 2011 modeling episode. The NH_3 concentration is subject to considerable variability, not only on a diurnal basis but also day to day. The variability predicted by the model is similar to the measured variability, although daily peaks predicted by the model are not aligned with measured peaks. This is understandable, in that the model incorporates variations caused by meteorological parameters but does not incorporate information on the timing of animal waste handling operations. Animal wastes are periodically distributed to fields using high-pressure sprays which can result in emissions of NH_3. However, information on the timing of this operation is not compiled for the inventory.

The daytime peaks for the model in Figure 4 are generally higher than the measured daytime peaks. This corresponds with the overprediction discussed above and shown in Table 1 and Figure 3. The overprediction at Clinton may be partially attributable to the location of the monitor at a boundary between an agricultural field and a forested area, which may result in the enhanced deposition and attenuation of transport from some directions. A review of deposition measurements by Schrader and Brummer (2014) indicates that deposition velocities are higher in forested areas than in agricultural areas.

The other available ground level measurement sites are more distant from the region of dense CAFO emissions, and measured concentrations at these sites are low. Atmospheric NH_3 concentrations at the three North Carolina AMoN sites were 1.3 ppbv or less; and concentrations of particulate NH_4^+ were 1.3 μg/m^3 or less. Table 1 shows that the model prediction was within the range of uncertainty at the Candor AMoN site in Central North Carolina and at Coweeta in the West.

The model underpredicted NH_3 at Beaufort, near the seacoast, by 81%. The magnitude of this error, 1.1 ppbv, was small in absolute terms, and a number of factors could have contributed to this underprediction. Ammonia emissions from the ocean could be contributing a small increase in NH_3 at the monitor (Paulot et al., 2015). Such emissions are not included in the model. The underprediction of NH_3 at Beaufort could also be attributable in part to an underestimation of NH_3 emissions from chemical fertilizer near the site, because the monitor site is adjacent to a large farm which alternates between growing corn and soybeans. Synthetic nitrogen fertilizer is used in the spring months when corn is cultivated (in odd-numbered

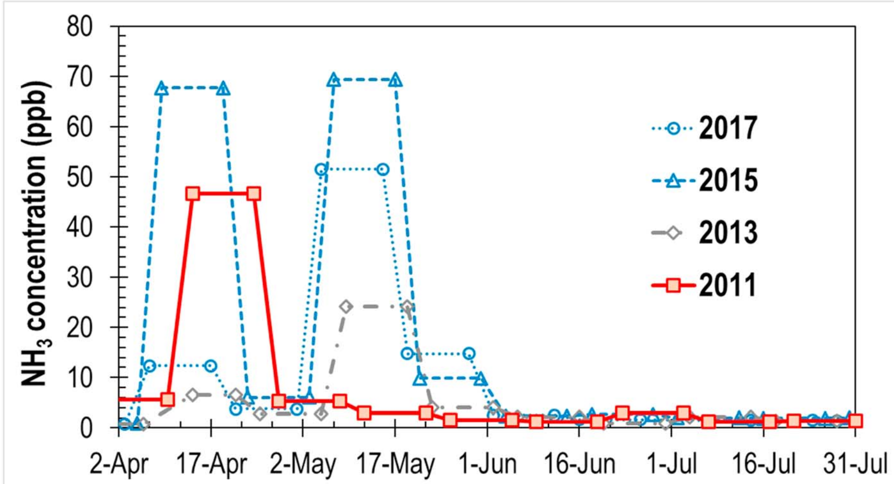

Figure 5. NH_3 associated with fertilizer application for a 2-year crop rotation at a large farm adjacent to the Beaufort Ammonia Monitoring Network site.

years), resulting in high measured atmospheric concentrations of NH_3 at the monitor—up to 70 ppbv on a 2-week average (see Figure 5). The spike in NH_3 is attenuated by July, but enhanced levels of nitrogen in the ground could still produce elevated concentrations on the scale of 1 ppbv. The bidirectional model gives an improved treatment of emissions from fertilized crops; however, the model does not currently address crop management and fertilizer practices at the model grid scale (Cooter et al., 2012).

In addition, the model overestimated airborne particulate NH_4^+ at Beaufort by 0.46 μg/m^3, an overprediction of 80% (Table 2). This overprediction may be indicative of an overestimation of the conversion of NH_3 to NH_4^+ at the Beaufort site. An error of 0.46 μg/m^3 in the production of NH_4^+ would correspond to a difference in NH_3 concentration of 0.65 ppbv or 60% of the NH_3 underprediction.

Table 2 compares predicted levels of airborne particulate NH_4^+ with corresponding measurements from CASTNET monitors collocated with the Beaufort site and the two other North Carolina AMoN sites. Another CASTNET site is located at Cranberry, in Western North Carolina. The measured concentrations of NH_4^+ were low, the highest being 1.3 μg/m^3 in Cranberry and Candor. Model predictions of NH_4^+ were within the range of uncertainty at all of the sites except Beaufort.

Table 3 assesses model predictions for the combination of NH_3 and NH_4^+ (total ammoniacal nitrogen) and for the partition between NH_3 and NH_4^+ at the three collocated AMoN and CASTNET sites—Beaufort, Candor, and Coweeta. In order to combine and compare the two species, both are expressed in terms of the mass concentration of their elemental nitrogen (NH_3-N and NH_4^+-N in μg-N·m^{-2}·s^{-1}). Table 3 shows that model predictions of total ammoniacal nitrogen are within 30% of measured values for all three collocated sites. This is well within the range of uncertainty.

Table 3 shows molar ratios of NH_3-N to NH_4^+-N derived from measurements data and model predictions at the three collocated monitor sites. At Candor and Coweeta, the measured ratios of NH_3-N to NH_4^+-N are less

Table 2
Clean Air Status and Trends Network Measurements of NH_4^+ in Fine Particulate Matter Compared With Model Predictions

Monitor site and location	Measurement (μg/m^3)[a,b]	Model with modified emissions inventory		
		Prediction (μg/m^3)[a]	Absolute bias (μg/m^3)	Normalized bias (%)
Beaufort (BFT142), 34.8846°N, 76.6207°W	0.57 ± 0.3	1.0 ± 0.7	0.46	80
Candor (CND125), 35.2632°N, 79.8365°W	1.3 ± 0.4	1.5 ± 0.9	0.20	15
Coweeta (COW137), 35.0605°N, 83.4305°W	1.1 ± 0.2	1.0 ± 0.6	−0.10	−9
Cranberry (PNF126), 36.1054°N, 82.045°W	1.3 ± 0.3	1.1 ± 0.7	−0.25	−20

[a]Average and standard deviation. [b]Five 1-week averages from 28 June through 2 August.

Table 3
Measurements and Model Predictions for the Combination of NH_3 and NH_4^+ and for the Partition Between NH_3 and NH_4^+

Monitor site and location	Total NH_3 and NH_4^+ as nitrogen (μg-N·m^{-2}·s^{-1})		Normalized bias (%)	Ratio of NH_3-N to NH_4^+-N (molar)	
	Measured	Modeled		Measured	Modeled
Beaufort (NC06), 34.8846°N, 76.6207°W	1.2 ± 0.3	0.94 ± 0.6	−21	1.7 ± 0.9	0.17 ± 0.2
Candor (NC26), 35.2632°N, 79.8365°W	1.5 ± 0.6	1.9 ± 0.9	27	0.45 ± 0.5	0.60 ± 0.5
Coweeta (NC25), 35.0605°N, 83.4305°W	1.1 ± 0.2	0.94 ± 0.6	−16	0.30 ± 0.1	0.22 ± 0.2

than one (0.45 ± 0.5 and 0.30 ± 0.1, respectively). Modeled ratios are also below one and are within the range of uncertainty in both cases. The Beaufort monitor stands out, in that the ratio of NH_3-N to NH_4^+-N is greater than one (1.7 ± 0.9). In contrast, the model predicts a ratio much lower than one (0.17 ± 0.2) for the Beaufort site. The high measured ratio of NH_3-N to NH_4^+-N at Beaufort compared to other sites suggests the presence of a local source of NH_3 emissions that is missing or underestimated in the model, such as the ocean emissions or fertilizer emissions discussed above. At the same time, the very low modeled ratio of NH_3-N to NH_4^+-N suggest that the model may also have overestimated the conversion of NH_3 to NH_4^+.

3.2. Model Predictions Compared With Satellite Retrievals

Satellite retrievals provide an opportunity to evaluate model performance at reproducing regional patterns in NH_3 loading. Figure 6 shows the spatial pattern atmospheric column NH_3 loading based on IASI retrievals (Version 2.2) for July 2011. The grid network is the same as the CMAQ modeling grid. In Figure 6, each grid value is actually an average of the retrievals for itself and its eight neighbors. This is done in order to increase the number of observations incorporated into each value. Table 4 compares average IASI retrievals of total column NH_3 loading (Version 2.2) for different geographic regions of the modeling domain with CMAQ predictions using the two emission inventory versions. Model-to-measurement comparisons are made for Sampson and Duplin counties, which have the highest swine population density, as well as for the eastern and western portions of North Carolina and the adjacent coastal waters. The model produced good agreement with IASI retrievals but showed some regional variation. Model predictions were 26% higher than IASI retrievals over the emission-dense area of Sampson and Duplin counties but comparable to IASI retrievals over the rest of the state. The differences are within the range of uncertainty of the model predictions and of the satellite retrievals. For comparison, the weighted average relative error of the Version 1 retrievals over Sampson and Duplin counties was 60%.

The model underpredicted the NH_3 loading in the offshore waters southeast of North Carolina by about 80%. This is most likely the result of fires which occurred in the coastal forests in late June and early July. The

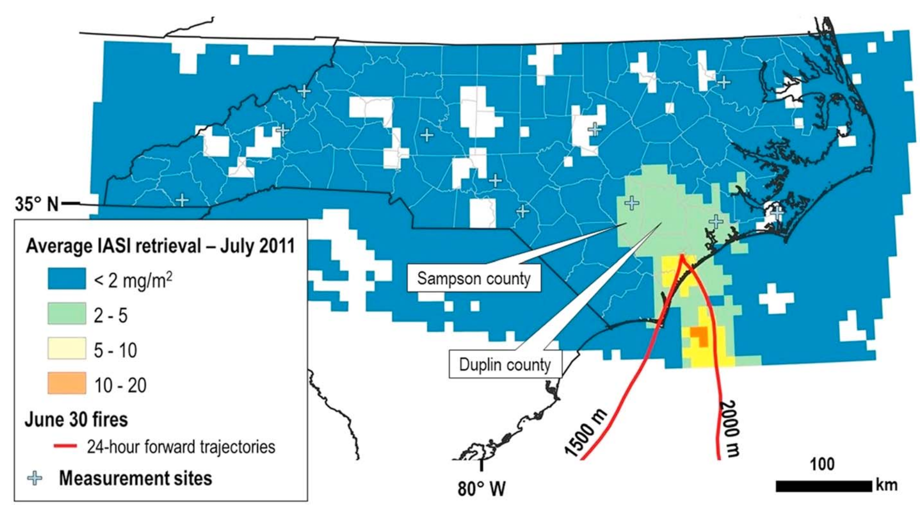

Figure 6. Spatial pattern of total atmospheric NH_3 loading based on average IASI retrievals for July 2011. IASI = Infrared Atmospheric Sounding Interferometer.

Table 4
IASI Retrievals of NH_3 (Version 2.2) in the Total Atmospheric Column Compared With Model Predictions

Region	IASI daytime retrievals Average (mg/m^2)	IASI daytime retrievals Standard deviation (mg/m^2)	IASI daytime retrievals Number of observations	Model prediction with modified emissions inventory Average (mg/m^2)	Model prediction with modified emissions inventory Standard deviation (mg/m^2)	Model prediction with modified emissions inventory Absolute bias (mg/m^2)	Model prediction with modified emissions inventory Normalized bias (%)
Sampson and Duplin counties	2.7	2.4	102	3.4	1.3	0.7	26
Eastern NC	1.2	3.4	1,030	1.2	1.3	<0.1	−2.1
Western NC	0.4	3.2	856	0.4	0.6	<0.1	−3.6

Note. IASI = Infrared Atmospheric Sounding Interferometer; NC = North Carolina.

bulk of these emissions occurred prior to the initiation of the model run (U.S. EPA, 2018b) and was not included in the initial conditions of the run. Figure 6 includes the locations of these fires, based on the satellite-based burn area measurement database from the Moderate Resolution Imaging Spectroradiometer (Giglio et al., 2009; Roy et al., 2008). The figure also shows 24-hr forward wind trajectories at the time of the fires (Rolph et al., 2017; Stein et al., 2015).

3.3. Model Predictions Compared With Deposition Measurements

Table 5 compares CMAQ predictions of wet NH_4^+ deposition with measured wet deposition at eight NTN sites across North Carolina. The table compares predictions and measurements of rainfall NH_4^+ concentrations, rainfall amounts, and deposition rates. In general, the model underpredicted rainfall NH_4^+ concentrations by up to 45%. With the exception of the Beaufort site, the model underpredicted the monthly wet NH_4^+ deposition by up to 61%. Model predictions for the Clinton Crop Research Station, in the region of highest emission density, are closest to the observed results, with a bias of −22% for the NH_4^+ rainfall concentration and −17% for the monthly wet NH_4^+ deposition.

At the Beaufort site, near the coast, the model overpredicted rainfall by almost 200%. As a result, wet NH_4^+ deposition was overestimated by 89% even though the concentration of NH_4^+ in rainfall was underestimated by 37%.

Dry deposition measurements for NH_3 and particulate NH_4^+ are rare. However, detailed measurements are available for NH_3 deposition at Finley Farm, an agricultural research facility in East Central North Carolina, in 2002. Because emissions and meteorological conditions would not be identical in the two years, the model predictions of NH_3 concentration and dry deposition flux would not be expected to directly match measured results for these parameters. However, the dry NH_3 deposition velocity can be expected to be comparable. (The dry deposition velocity is the ratio of NH_3 deposition flux to the atmospheric concentration.) Table 6 shows CMAQ predictions of the dry deposition rate for NH_3, along with the dry deposition velocity at the Clinton monitor site and the Finley site. The table also compares NH_3 dry deposition velocities predicted

Table 5
National Trends Network Measurements of Wet Deposition Compared With Model Predictions

Monitor site and location	Rainfall NH_4^+ concentration Measured (mg/L)	Rainfall NH_4^+ concentration Modeled (mg/L)	Rainfall NH_4^+ concentration Model bias (%)	Rainfall amount Measured (cm)	Rainfall amount Model (cm)	Rainfall amount Model bias (%)	Total deposition (July 2011) Measured (kg/ha)	Total deposition (July 2011) Model (kg/ha)	Total deposition (July 2011) Model bias (%)
Clinton Research Station (NC35), 35.0258°N, 78.2783°W	0.70 ± 0.45	0.55 ± 0.13	−22	7.0	7.4	6	0.49	0.41	−17
Beaufort (NC06), 34.8846°N, 76.6207°W	0.29 ± 0.41	0.19 ± 0.06	−37	5.7	17.0	197	0.17	0.32	89
Lewiston (NC03), 36.1325°N, 77.1708°W	0.49 ± 0.36	0.32 ± 0.06	−35	13.8	10.0	−27	0.68	0.32	−53
Hofmann Forest (NC29), 34.825°N, 77.3228°W	0.52 ± 0.48	0.39 ± 0.17	−24	17.2	13.0	−25	0.89	0.51	−43
Finley Farm (NC41), 35.7288°N, 78.6802°W	0.48 ± 0.13	0.26 ± 0.02	−45	6.8	7.6	13	0.32	0.20	−38
Jordan Creek (NC36), 34.9705°N, 79.5281°W	0.37 ± 0.13	0.27 ± 0.08	−26	9.8	5.6	−43	0.36	0.15	−58
Piedmont Research Station (NC34), 35.697°N, 80.6225°W	0.31 ± 0.16	0.23 ± 0.18	−25	19.4	10.2	−48	0.60	0.23	−61
Mt. Mitchell (NC45), 35.7353°N, 82.2861°W	0.27 ± 0.11	0.27 ± 0.14	0	23.6	15.1	−36	0.64	0.41	−36

Table 6
Model Predictions of Dry Deposition Compared Measurements for the Findley Farm Site in 2002

	Concentration (ppbv)	Deposition flux ($\mu g \cdot m^{-2} \cdot s^{-1}$)	Model deposition velocity (cm/s)	Deposition velocity from Phillips (cm/s)	Difference (%)
Clinton site					
Day	15.4 ± 9.4	0.172 ± 0.105	1.6 ± 0.5		
Night	6.6 ± 12.1	0.036 ± 0.045	0.8 ± 0.5		
Average	11.3 ± 10.8	0.110 ± 0.080	1.4 ± 0.5		
Finley site					
Day	1.4 ± 1.2	0.016 ± 0.009	1.7 ± 0.9	3.9 ± 2.8	−56
Night	1.1 ± 0.9	0.005 ± 0.004	0.7 ± 0.3	0.76 ± 1.7	−10
Average	1.2 ± 1.1	0.011 ± 0.007	1.3 ± 0.5	3.0	−56

by the model for Finley Farm with dry deposition velocities measured by Phillips et al. (2004) at Finley Farm in 2002. The table shows that deposition velocities predicted by the model for Finley Farm and the Clinton site are similar (1.3 ± 0.5 cm/s for Finley Farm and 1.4 ± 0.5 cm/s for Clinton). In addition, the nighttime deposition velocity predicted by the model for Finley Farm is similar to the value measured by Phillips in 2002 (0.7 ± 0.3 cm/s for the model compared with 0.76 ± 0.5 cm/s for the Phillips measurement). However, the daytime deposition velocity predicted by the model is less than half of the value measured by Phillips (1.7 ± 0.9 cm/s for the model compared with 3.9 ± 2.8 cm/s for the measurement).

3.4. Analysis of Model Bias in Relation to Previous Studies

Gilliland et al. (2006) conducted a CMAQ inverse modeling analysis using wet NH_4^+ deposition measurements to evaluate the 2001 U.S. EPA emission inventory of NH_3 for the continental United States. This study found that annual emissions estimates were found to be reasonable on average but that the emissions inventory was about 17% too low in the July–August time frame. Zhu et al. (2013) carried out inverse modeling with the GEOS-Chem model using satellite retrievals of ambient NH_3 from the Tropospheric Emission Spectrometer on the Aura satellite and ground level AMoN measurements between 2006 and 2009 for the Continental United States. The study found that the emissions inventory for NH_3 appeared to be an underestimate, especially in the Western United States.

Butler et al. (2014) found that CMAQ predictions of atmospheric NH_3 were 8% to 60% lower than measured values for the Susquehanna River Watershed of New York and Pennsylvania in 2008 and 2009. Kelly et al. (2014) evaluated CMAQ predictions of NH_3 and NH_4^+ in airborne particulate matter as part of the California Research at the Nexus of Air Quality and Climate Change campaign in May and June of 2010. Predictions of NH_4^+ were close to measured values; however, the model underpredicted NH_3 in agricultural regions and did not capture the large variations in measured NH_3.

Using aircraft-based measurements, ground level measurements, and Tropospheric Emission Spectrometer satellite retrievals, Battye et al. (2016) found that the NAQFC CMAQ model underpredicted atmospheric NH_3 by 33% to 76% in an agricultural region of Northeast Colorado in the summer of 2014, although predictions of particulate NH_4^+ were close to measured values. Bray et al. (2017) found that the NAQFC CMAQ model underpredicted atmospheric NH_3 by 58% for the San Joaquin Valley of California in the summer of 2010.

In general, the previous studies cited above have shown underprediction of NH_3 in the summer in other agricultural regions of the United States. This current study differs, showing an overprediction of NH_3 at Clinton, within the area of densest animal population. However, it must be noted that these studies encompass different animal populations. Animal production in Eastern North Carolina was dominated by hogs in 2011, with an increasing contribution from poultry operations since then. Ammonia emissions in the Susquehanna Valley are dominated by poultry operations; while emissions in California and Northeast Colorado are dominated by cattle (U.S. EPA, 2015).

Emissions estimates for animal waste are subject to sources of potential error and bias which can vary by animal category and by geographic region. Agricultural census data are available only available for broad categories of animals, and emission factors are not available for all categories and situations. For instance,

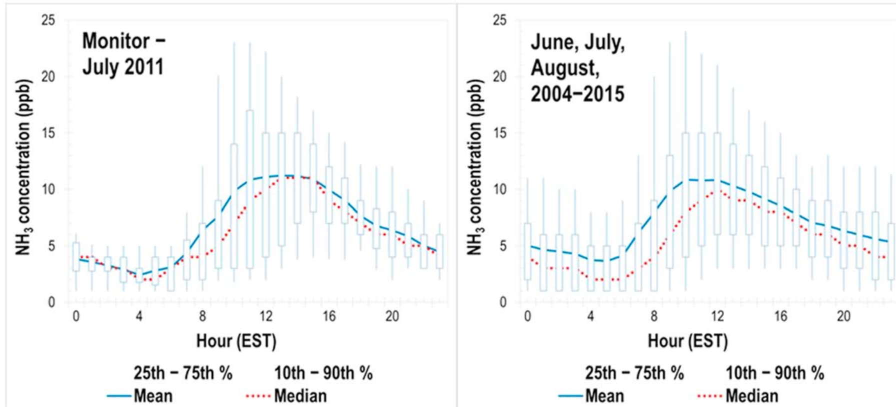

Figure 7. Comparison of the monitored NH_3 concentrations during July 2011 with longer-term monitoring data. Solid line and dotted line show the mean and median, respectively. Vertical box shows 25th and 75th percentiles, and vertical lines show 10th and 90th percentiles.

all swine are lumped into a single category, without any differentiation by animal size (USDA, 2019). However, an animal's rate of waste generation changes substantially as the animal grows. The age distribution of swine can vary considerably for operations in North Carolina (NCDEQ, 2017). An operation with a higher proportion of younger animals would have lower emissions on a per animal basis than an operation with a higher proportion of older animals. For the 2011 NEI and prior inventories, emission factors did not discriminate between cattle on pastures and cattle in feedlots. Similarly, NEI emission factors to date have not differentiated between different types of waste management systems. Waste management techniques vary for different regions of the country. Lagoon-based management systems are dominant for hog operations in North Carolina, while pit systems are used in some other regions. Thus, there are a number of reasons why emissions for some animal categories and regions could be understated, while emissions for other animal categories and regions could be overstated.

3.5. Placing the July 2011 Modeling Period in Context With Longer-Term Measurement

The model overprediction in the region of dense animal population raises the question of whether the July 2011 study period is anomalous in some way. Figure 7 compares the pattern of NH_3 concentrations measured for July 2011 with the pattern of concentrations measured for all summer months (June, July, and August) during the 11-year time frame in which the Clinton monitor was operating. The mean, median, and 10th, 25th, 75th, and 90th percentiles are shown for both data sets. The figure shows that the average diurnal pattern in July 2011 is similar to that in other summer months and that the range of concentrations measured is similar.

Figure 8 shows long-term trends in wet deposition at the Clinton monitoring site, along with long-term trends in swine and poultry populations in Sampson County, where the site is located. In order to account for differences in waste generation between animal types, the populations are expressed in animal units (a.u.). Animal populations were derived from annual surveys and 5-year Censuses carried out by the U.S. Department of Agriculture (USDA, 2019). Animal unit factors for swine, turkeys, and chickens were taken from the Illinois Animal Management Facilities Act, which provides a commonly used list of a.u. factors (Illinois Statutes, 1996). The figure shows that there has been an increase in wet NH_4^+ deposition since measurements were initiated in 1978. The increase was rapid prior to the Year 2000, corresponding to substantial increases in the population of hogs raised in the counties surrounding the Clinton monitor. The rate of increase slowed substantially after 2000, corresponding with the adoption of a moratorium by the state government on permitting of new hog production facilities.

3.6. Impact of Meteorological Conditions on Model Bias

The model overprediction at the Clinton monitor also raises the question of whether the model was correctly predicting the wind distribution and precipitation at the monitoring site. Figure 9 compares the distribution of wind speed and direction measured at the nearby Clinton airport with those predicted by the model for the

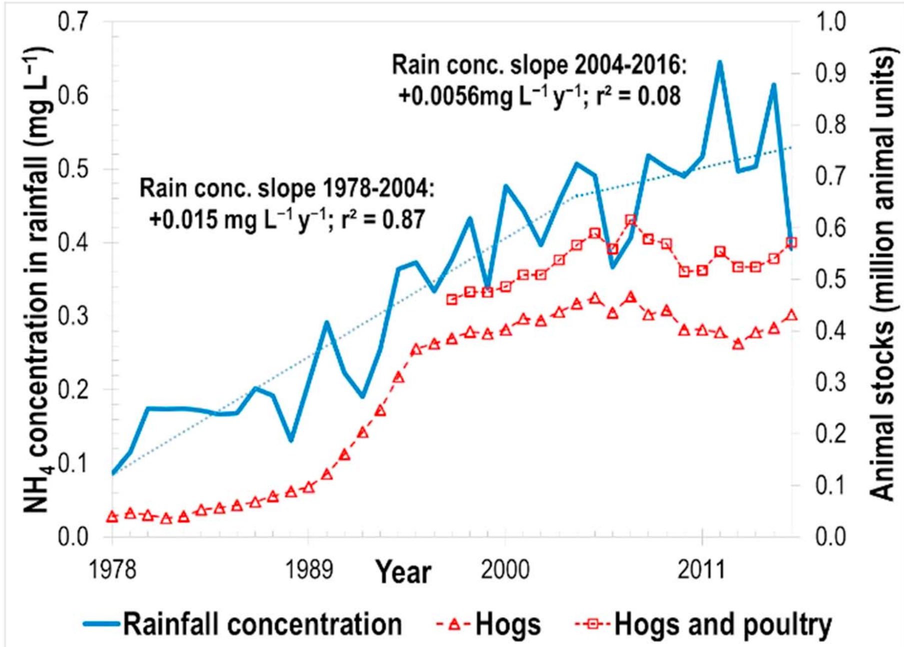

Figure 8. Long-term trends in rainfall NH_4^+ concentration at the Clinton monitoring site compared with population trends for swine and poultry in Sampson County. (Animal populations are expressed in a.u. to account for differences in mass and waste generation between species. Swine weighing over 25 kg are treated as 0.4 a.u., swine under 12 kg are treated as 0.03 a.u., hens and broilers are treated 0.01 a.u., and turkeys are treated as 0.02 a.u. The census for poultry was incomplete prior to 1997.) a.u. = animal unit.

airport location. Meteorological observations for the Clinton airport (WBAN 03727) were obtained from the NOAA National Centers for Environmental Information (NCEI, 2017). Figure 10 shows the influence of wind speed and direction on the NH_3 concentration measured at Clinton and predicted by the model during the hours of highest NH_3 concentrations from 10 A.M. to 6 P.M. local time. The figure also shows the ranges of observations and predictions for the different wind conditions, as reflected by geometric standard deviations.

Figure 9 shows that the model did a reasonable job of predicting the distribution of wind speeds and directions at Clinton during July of 2011. The model predicted a higher frequency of wind from the Northeast than was observed. However, Figure 10 shows that model NH_3 predictions for northeast winds are not higher than average model predictions under other conditions.

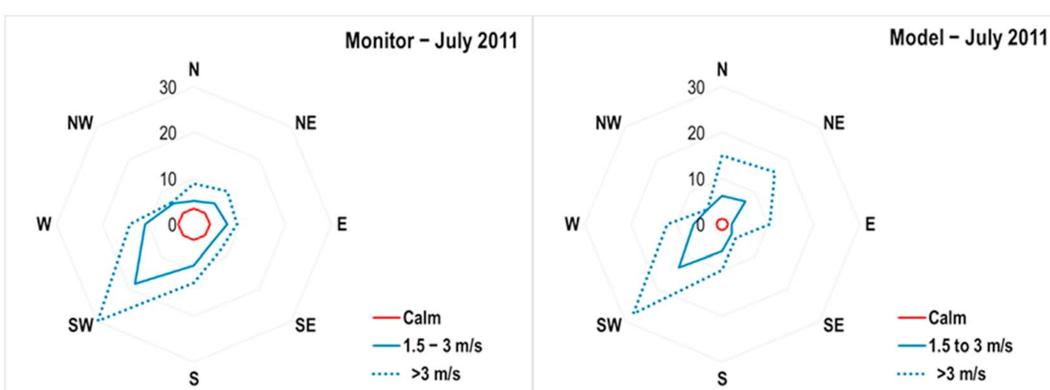

Figure 9. Comparison of the measured wind rose for Clinton with the modeled wind rose.

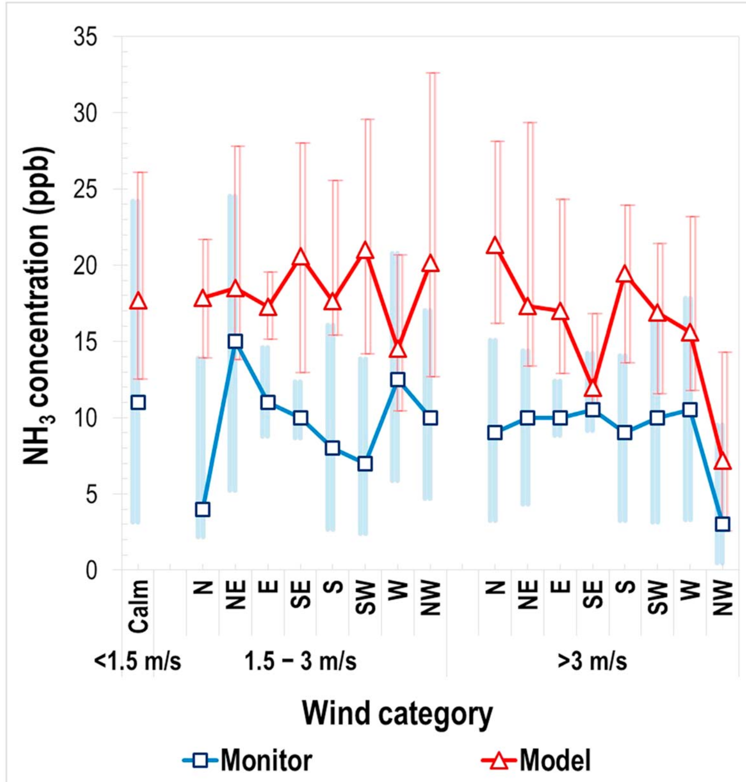

Figure 10. Dependence of measured and predicted NH_3 concentrations on wind speed and direction during the hours of highest NH_3 concentrations from 10 A.M. to 6 P.M. (Values reported are medians, and error bars reflect the geometric standard deviations of the observations within each category.)

The largest overprediction shown in Figure 10 is for northerly winds, where the monitored concentrations were depressed while the modeled concentrations were not. This corresponds with the location of a forested area immediately north of the monitor site, which may have attenuated NH_3 transported from that direction. However, this wind condition was relatively infrequent. It must also be noted that uncertainty is large for the average NH_3 measurement or prediction under any particular wind condition.

In the timeline of daily average NH_3 concentrations (Figure 4), the 10 and 11 July are somewhat anomalous in that the observed average NH_3 concentrations noticeably exceeded predictions for those days. Rain was not predicted or observed on either day. On both days, prolonged periods of calm conditions were observed in the afternoon, but these calm conditions were not predicted by the model. In fact, the model generally predicted fewer periods of calm conditions than were observed (Figure 9). Although calm conditions do not produce a noticeable increase in the median NH_3 measurement, the range of measured concentrations under these conditions is broad.

Rain was measured between the hours of 10 A.M. and 6 P.M. on 7 days of the study period, compared with 8 days predicted by the model. However, the model predicted significantly less rain than was observed. For the overall study period, the model predicted a total of 4.1 cm of rain, compared to a total of 8.0 cm measured. The impact of rain on model bias is difficult to quantify, since rain may last for only a few hours a day, and the rain can occur during different phases of the diurnal NH_3 emission cycle. Observed concentrations of NH_3 were 31% lower on average during rainy periods than periods without rain, while modeled concentrations were only 15% lower during rainy periods. This difference is manifested in the period from 24 July through 26 July, during which rain was observed and also predicted by the model. The model predicted only 1 cm of precipitation during this, whereas 4.1 cm was measured. Both the observed and modeled NH_3 concentrations were below their averages, but the observed concentration fell lower than the modeled concentration. Notwithstanding the impacts of rain, the model still overestimated NH_3 by an average of 63% for days when rain did not occur.

Both the measured and modeled concentrations showed large variations during the course of the month. However, these variations may have different sources. The model concentration depends on the density of emissions in the upwind direction, the wind speed, the degree of mixing, and the rate of deposition. The monitored concentration varies with all of these parameters and also varies with short-term changes in emissions. The model takes into account the impacts of temperature on emissions but does not incorporate information on the timing of waste management operations. Actual emissions are strongly affected by short-term waste handling processes, such as periodic waste spraying. Figure 4 showed that modeled concentrations generally exceeded measured concentrations, with a few exceptions where measured concentrations spiked to levels exceeding the modeled values.

4. Conclusions

Even with a 42% downward adjustment of NH_3 emissions from CAFOs in the study region, the NAQFC CMAQ model overpredicted atmospheric NH_3 in July 2011 by 60% at the Clinton continuous monitoring site. The average concentration measured by the monitor was 6.6 ppbv for July 2011, while the average predicted by the model was 10.5 ppbv. The monitoring site is located in Sampson County, in a region of dense NH_3 emissions from swine operations. The model bias at the Clinton site was reduced by the use of CAFO permit information to improve the spatial allocation of NH_3 emissions. In a standard CMAQ model run, NH_3 emissions are calculated at the county level and then allocated to model grids based on land use. In the current study, emissions were allocated to model grids using CAFO permit information. This spatial allocation modification reduced emissions by 41% in the model grid containing the Clinton monitor. Therefore, model bias without the spatial adjustment could be expected to be up to 41% higher than the current overprediction (60%). It must also be noted that the current study did not include a recently developed bidirectional exchange algorithm for NH_3. Implementing this algorithm may increase NH_3 concentrations in source regions by 10% or more (Bash et al., 2013; Cooter et al., 2012).

Methodologies for calculating agricultural NH_3 emissions are changing for the 2014 NEI, as the U.S. EPA is moving to a more refined emission process modeling approach for animal wastes (McQuilling & Adams, 2015). With this new approach, there is the potential to estimate emissions based on the volume of waste handled, rather than the animal population. This would allow the incorporation of the age distribution of animals into the inventory. In addition, inventory process models can take into account different waste management practices, as well as approaches to mitigate emissions. However, changes to the emissions inventory process will be needed in order to collect and incorporate this information.

This current study is the first to use hourly NH_3 concentrations from the Clinton monitor to evaluate ground level predictions by the CMAQ model, providing new information on the performance of the model for reproducing the diurnal pattern of NH_3 concentration. Previous studies have used passive monitors with a temporal resolution of at least 1 week. The diurnal pattern predicted by the model was similar to that measured by the continuous monitor, with both showing elevated concentrations in the midafternoon and lower concentrations at night.

The other available ground level measurements are more distant from the region of dense CAFO emissions. Atmospheric NH_3 concentrations at the three North Carolina AMoN sites were 1.3 ppbv or less; and concentrations of particulate NH_4^+ were 1.3 $\mu g/m^3$ or less. With the exception of one monitor, model predictions at these sites were within the range of uncertainty. The exception was the Beaufort monitor, near the seacoast, where the model underpredicted atmospheric NH_3 by 1.1 ppbv and overpredicted particulate NH_4^+ by 0.4 $\mu g/m^3$. The underprediction of NH_3 may be attributable to the location of the Beaufort monitor in close proximity to a fertilized corn field or to NH_3 emissions from the ocean nearby. Total column concentrations of NH_3 predicted by the model were 26% higher than IASI Version 2.2 retrievals for the emission-dense area of Sampson and Duplin counties; however, this difference is within the range of uncertainty of the satellite retrievals. Predicted column concentrations were in good agreement with IASI retrievals over the rest of North Carolina.

It must be noted that the Clinton monitor is collecting data from a single point within a 12-km model grid. The measured concentration at this point may not be representative of the entire grid. The model bias at Clinton was largest for winds from the north, corresponding to the direction of a forested area very close to the monitor. This results in the attenuation of NH_3 transported from this direction. Large temporal variations in emissions also complicate any comparison of modeled versus monitored concentrations. The emissions inventory takes into account the influence of temperature on emissions but does not take into account the impacts of short term waste management practices. For instance, swine waste is periodically distributed to croplands using a high pressure spray. Information is not generally available on the timing of this operation.

The overprediction at Clinton may also be attributable, at least partially, to the broader underprediction of wet and dry deposition across the study region. The model underpredicted wet deposition of NH_4^+ and dry deposition of NH_3. The concentration of NH_4^+ in rainfall was underpredicted by 22% at Clinton and up to 45% at Finley Farm in Central North Carolina. The model underpredicted monthly wet deposition NH_4^+ by 17% at Clinton and by up to 61% at the Piedmont Research Station in Central North Carolina. The predicted daytime dry NH_3 deposition velocity was 57% less than that measured by Phillips et al. (2004), and the overall dry NH_3 deposition velocity was 56% less than that measured by Phillips et al. (2004).

The State of North Carolina discontinued continuous monitoring at the Clinton site in 2015, and the continuous monitor has not been replaced by a passive monitor. This means that currently, the Beaufort AMoN site is the nearest NH_3 monitor to the region of densest animal populations in Sampson and Duplin counties. Ongoing monitoring is needed for NH_3 within the region of densest animal populations.

Additional measurements of wet and dry depositions are needed, especially in the region of dense animal population around Clinton. The measurements of dry deposition velocity at Finley Farm are valuable but are dated (2002). In addition, the Finley Farm location is about 50 km northwest of the region of dense animal population. Additional dry deposition measurements in areas such as the Clinton region would not only help to improve model estimates of NH_3 but would also allow a more accurate assessment of nitrogen deposition to sensitive ecosystems in the region. The animal production region around Clinton is located within 50 km of the Albermarle-Pamlico Estuary, the Neuse River Estuary, and other river estuaries. Trend analysis indicates that the wet deposition of NH_4^+ at Clinton is increasing. Understanding and prediction of such trends in reactive nitrogen species are important to assess the impacts of nitrogen deposition on terrestrial and aquatic ecosystems.

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