Carnegie Mellon University

CARNEGIE INSTITUTE OF TECHNOLOGY

THESIS

SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS

FOR THE DEGREE OF Doctor of Philosophy

TITLE Ammonia Emissions from Livestock in the United States:

From Farm-Level Models to a New National Inventory

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ACCEPTED BY THE DEPARTMENTS OF

Civil and Environmental Engineering

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APPROVED BY THE COLLEGE COUNCIL

Vijayakumar Bhagavatula — DEAN — February 23, 2016 — DATE

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in

CIVIL AND ENVIRONMENTAL ENGINEERING

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February, 2016

ACKNOWLEDGEMENTS

I am sincerely grateful to have spent the last five years at Carnegie Mellon University doing research with the Center for Atmospheric Particle Studies, and I am so glad to have met not only colleagues but friends in my research group. Special thanks to my fellow CAPS modelers. I cannot imagine having completed my studies anywhere else.

Many thanks to the members of my committee, Neil Donahue, Spyros Pandis, and Mitch Small, for all of the advice and insights provided throughout the past five years. Their help and mentorship was truly invaluable. Special thanks to my advisor, Peter Adams for not only advice, but for the flexibility and encouragement you gave me as I completed my work.

Many thanks to my family and to the friends I have made in Pittsburgh that have become like a second family to me. I am thankful for the support of my parents who always encouraged me to “just do my best” and for my sister who always competed with me in school and pushed me to do even better.

Finally, my thanks to my husband, JP, for his support, for listening to me practice all of my research talks, and for his patience over the more than five years that we lived in different states; I can’t imagine having a better partner to share the graduate school experience with and am eternally grateful for his encouragement as we each pursued our degrees.

This research was supported by EPA Agreement Number: RD-83454901-0.

ABSTRACT

The role of nitrogen as an environmental pollutant has long been established, but the significance and amount of ammonia emitted from livestock production has not been well-characterized in the United States. In order to better characterize the impacts of ammonia as an air pollutant and its role in the formation of fine particulate matter, we have used a semi-empirical process-based model to estimate ammonia emissions from beef cattle, swine, layer chickens, and broiler chickens in the United States. The semi-empirical model is used so that the ammonia emissions can be simulated using a mass balance on nitrogen through the farm, tracking nitrogen from housing, storage, and application of manure, in addition to being constrained by a number of tuned parameters which ensure that the resulting emission factors are realistic. In addition, model inputs, including manure pH, manure volume, manure nitrogen content and manure dry matter content are taken from the existing literature. These previous studies were the first means by which the FEMs were evaluated, with the FEMs able to capture 20-70% of the variability in reported emissions. Next, observations from the National Air Emissions Monitoring Study were used to further test and evaluate the model, including the evaluation in its performance to characterize seasonal as well as day-to-day variability in emissions of ammonia; here again, seasonal variability was well-characterized with slightly more mixed results for daily variability in ammonia emissions. Then, emissions from these single farms were summed over the entire populations of cattle, swine and poultry in the US, using data from the National Climate Data Center and USDA National Animal Health Monitoring Survey for appropriate meteorology and manure management practices in different locations. Emissions total 1.7 Tg from our model: 723 Gg from swine, 435 Gg from beef, 202 Gg from dairy, and 357 Gg from poultry. These inventories were compared to the National Emissions Inventory which estimates 1.9 Tg of ammonia emissions from livestock in 2011. This work has demonstrated the model's ability to characterize the variability in ammonia emissions from livestock and can be used to understand the air quality impacts of this variability.

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CHAPTER 1: INTRODUCTION

1.1 Ammonia: a critical air pollutant

Ammonia, NH_3, causes numerous negative impacts on the environment and human health. Ammonia is also an irritant and corrosive at high concentrations. It contributes to the eutrophication of ecosystems through nitrogen deposition. Acute exposures to high levels of ammonia are associated with severe, permanent damage to the lungs (Leduc et al., 1992). Through the advent of the Haber process that allows for the industrial fixing of nitrogen into reduced forms, large amounts of reduced nitrogen are environmentally available and "will… cause damage to environmental services at local, regional, and global scales due to a large increase in reactive N load in the environment" (Galloway et al., 2008). Effects of excess nitrogen on ecosystems include "water pollution and reduced biological diversity" (American Chemical Society, 2011). Perhaps most importantly, ammonia emissions result in the formation of fine particulate matter (PM) which causes damage to human health and the environment (Draaijers et al., 1989; Erisman et al., 1998; Krewski et al., 2003; Pope and Dockery, 2006).

Specifically, fine PM is comprised of a mixture of organic and inorganic components, and the key inorganic components of PM are sulfate, nitrate, and ammonium (from ammonia emissions) (USEPA, 2004). These three species react in the atmosphere to form fine PM, also called $PM_{2.5}$ (particles with an aerodynamic diameter of 2.5 microns or less), and while sulfate and nitrate concentrations have decreased over the past few decades as a result of the sulfur dioxide cap and trade regime implemented by the US EPA and tighter regulations of NOx emissions, ammonia emissions have not been reduced, and are even expected to increase as agricultural production has intensified (National Research Council: AdHoc Committee on Air Emissions from Animal Feeding Operations, 2003; Rico, 1995).

In the formation of fine inorganic PM, ammonium first reacts to neutralize atmospheric sulfate; any leftover or free ammonia reacts with nitrate to form ammonium nitrate, and this reaction can be especially important during winter because this reaction is more thermodynamically favorable at lower ambient temperatures (Seinfeld and Pandis, 2012; Stanier et al., 2012). Because of the sensitivity of ammonium nitrate formation to ammonia emissions, especially during cold temperatures, it is vital to understand how ammonia emissions vary throughout the course of the year. Furthermore, ammonia is important in terms of both the mass of particulate matter in the atmosphere, described above, but also in relation to the formation of new particles. In ternary nucleation (a type of new particle formation), ammonia, sulfuric acid, and water form new particles, especially in lower altitudes where there is more ammonia present. This has been proposed as a mechanism (Kulmala, 2003) as well as observed in the laboratory (Ball and Hanson, 1999; Yu, 2006) and the atmosphere (Weber et al., 1999).

Currently, the National Ambient Air Quality Standards directly regulate levels of pollutants by specifying the maximum allowable ambient concentrations of six important air pollutants including particulate matter (PM) of both coarse and fine (aerodynamic diameter less than 2.5 μm) sizes (USEPA, 2014). Since ammonia contributes to the formation of fine PM it is possible that in order to control fine PM levels, ammonia emissions will need to be limited in the future. Alternatively, the regulation of ammonia could also be pursued under the Community-right-to-know Act which currently requires the reporting of releases of ammonia of greater than 100 pounds per 24 hours (45 kg per 24 hours) to the EPA for (industrial) point sources. Though agricultural sources are currently not included in this statute, many of the practices in common use, particularly the production of animals at concentrated animal feeding operations (CAFOs) more closely resemble point sources than the area sources that they are currently considered to be. If this regulation is implemented, large agricultural operations would be treated as industrial point sources. Currently, the implementation of this statute would be difficult because of limited agricultural monitoring and lack of data about the emission factors for different types of livestock (Donham et al., 2002; Martin, 2008; NIOSH et al., 1992). Even with current projects, including recent large measurement campaigns (i.e. National Air Emissions Monitoring Study, NAEMS), the entire range of variables that can be encompassed in the management and production of beef cattle, poultry, and swine in the United States are still not measured.

As mentioned above, the key ammonia emissions sources in the United States are from agriculture, specifically from the production of livestock. Ammonia from cattle, swine, and poultry accounts for 55% to 75% of total nation ammonia emissions (Battye et al., 1994, 2003; Galloway et al., 2008), and the characterization and estimation of emissions from these sectors are addressed in the following chapters.

Much of the current research on ammonia focuses on the monitoring of emissions from animal agriculture, and trying to estimate per animal emission factors under a variety of meteorological conditions and differing management practices as emissions can vary greatly based on differences in temperature, wind, precipitation and manure management practices. Figure 1.1a shows the typical contributions of various sectors to the total United States ammonia emission inventory, while Figure 1.1b displays animal type contributions to livestock ammonia emissions in the United States.

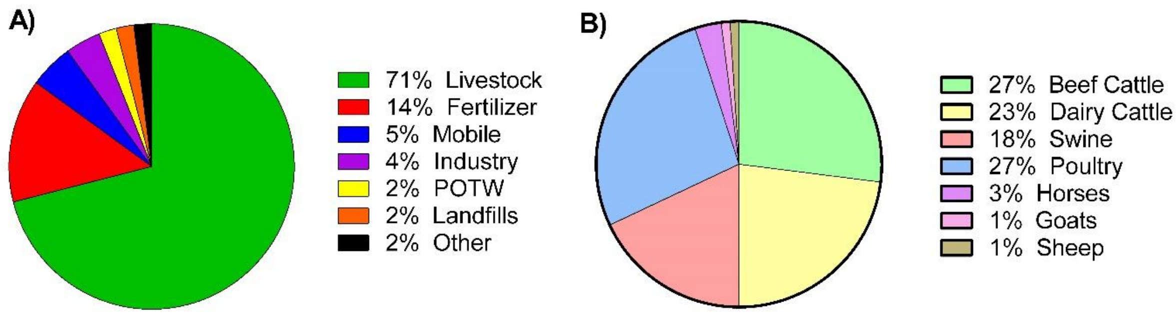

Figure **1.**1. (a) all US Ammonia Sources and (b) Livestock ammonia sources in the US (Roe et al., 2004)

According to recent inventories, approximately 70% of US ammonia emissions are from livestock production. Of that fraction, 95% of the emissions come from the following sources: 18% from swine, 27% from beef cattle, 23% from dairy cattle, and 27% from poultry, which includes both broiler chickens (produced for meat) and layer chickens (egg-producers).

1.2 Ammonia Emissions: Measurements and Sources of Variability

There are significant limitations to the ammonia emissions data available for beef, swine and poultry in the United States because there are limited resources available for the measurement of ammonia emissions. There is no way for emissions for each type of animal to be monitored under every possible condition and for every farm configuration since ammonia emissions depend on meteorology, management practices used, nitrogen in the feed and manure characteristics.

Higher temperatures result in an increase in the volatility of the ammoniacal nitrogen and lead to greater ammonia emissions. Higher wind speeds reduce surface resistance to mass transfer causing higher ammonia emissions. Finally, precipitation inhibits ammonia emission because it causes greater amounts of ammonia to infiltrate into the soil during either manure storage or application. Differences in management practices from farm to farm results include housing type, ranges and sources of crude protein in animal diet, manure storage time and conditions, frequency of pen or house-cleaning (cleaning is often associated with a burst of ammonia emissions), and methods of manure application also contribute to ammonia emissions variability.

Quantitatively, these differences in practices have been estimated to result in a range of annual emission factors from dairy cattle from 13 kg NH_3/year to 55 kg NH_3/year (Pinder et al., 2004b) and a range of daily emission factors from beef in the literature ranging from ~30 g NH_3/head/day to more than 200 g NH_3/head/day. For swine, similar ranges in emissions have been observed, with the greatest variability seen for swine manure storages which range from <1 g NH_3/head/day to more than 100 g NH_3/head/day (during the National Air Emissions Monitoring Study (NAEMS) particularly under warm, dry conditions). Poultry emissions also show a wide range, but are less driven by environmental conditions than those from dairy, beef or swine; layer emissions range from <100 g/AU/day to more than 500 g/AU/d (where AU = animal unit or 500 kg live animal weight). Broiler emissions depend strongly on animal age and can range from <100 g/AU/day to more than 1500 g/AU/day.

Published emission factors in the literature and from large scale monitoring campaigns (like the National Air Emissions Monitoring Study, NAEMS) only capture a small fraction of the meteorology, manure management practices, and manure characteristics that exist throughout the United States. Therefore, in order to estimate the ammonia emissions from farms which have not been monitored, we need a way to model ammonia emissions based on their meteorology and farm management practices.

1.3 Process-Based Models as a tool for estimating varying emissions

Traditional methods of developing emission inventories have relied on single emission factors which do not take into account meteorology or practices and how they differ across locations. Newer inventories may take into account empirical models that represent ammonia emissions as a function of a single variable, like temperature, in order to characterize some of the observed variability in emissions from different animal types. This approach has been employed for manure storages and broiler houses by EPA (USEPA-OAQPS, 2012) for the NAEMS farms and in the development of the most recent National Emission Inventory (NEI2011v2) (USEPA-OAQPS, 2015a). However, there are serious limitations to these methods as these empirical models are also often limited in their applicability in terms of conditions or locations for which their use is appropriate. An alternative to these purely empirical approaches is the process-based model.

Process-based models characterize variability in ammonia emissions by conducting a mass balance on the system, tracking a particular species, in this case ammoniacal nitrogen, throughout the entire system, breaking the production process into different stages to characterize all the emissions processes. This mass balance also describes emissions as a function of meteorological parameters and management practices. As stated, our farm emissions models are semi-empirical as they are based on a nitrogen mass balance with inputs of meteorological parameters and management practices to obtain the desired output of ammonia emissions as a function of time but will also be constrained through the use of tuned parameters to ensure agreement with previously reported ammonia emission factors. Our farm emission model (FEM) approach allows use to evaluate the model for consistency with measured emission factors, maintain consistency by tracking the actual nitrogen available for emission (rather than as a function of an environmental variable), and estimate uncertainty in our model's estimates of ammonia emissions, producing seasonally variable and daily variable. Furthermore, we can use these farm-level models to construct a temporally and spatially variable ammonia emissions inventory for swine, beef, dairy, layer, and broiler production in the United States. These FEMs are evaluated for both seasonal and daily performance for beef, swine, poultry and dairy ammonia emissions in Chapters 2 and 3.

1.4 A spatially and temporally variable livestock ammonia emissions inventory for the United States

As stated above, previous ammonia emissions inventories used a simple emission factor approach in their construction, where each source of emissions had an emission factor and an activity level, so emissions were simply calculated as shown in the equation below.

$$Emissions\ \left(\frac{kg}{y}\right) = Emission\ Factor\ (EF)\left(\frac{kg}{animal * year}\right) \times Activity(\#\ of\ animals)$$

The current NEI (National Emission Inventory) calculates the emissions for each source of agricultural ammonia as a function of the daily average temperature for a particular location; however, this does not capture all of the variability in ammonia emissions because it does not account for differences in emissions caused by other meteorological factors or differences in regional manure management practices.

The daily-resolved, county-level emission inventory described in Chapter 4 takes into account not only the effects of temperature, but also wind speed, precipitation, and the regional distribution of manure management practices for each animal type. By developing such a highly-resolved ammonia emissions inventory for livestock, we hope to be able to better estimate the effects of these emissions on other air quality problems, especially PM concentrations. Based on these results, we can begin to better understand the impacts of animal agriculture.

1.5 Inventory evaluation and data robustness

After the creation of a new inventory for ammonia emissions from beef, swine and poultry, we will need to evaluate our inventory. The first step of this process is discussed in Chapter 4 and is done through the comparison of the annual total emissions for swine, beef, dairy, layer, and broiler production for 2011 to the same totals produced by our FEM-based approach. Additional future evaluation will be done by implementing our process-based inventory in a chemical transport model and comparing the concentration data produced to data available from the National Atmospheric Deposition Program (NADP) and the Ammonia Monitoring Network (AMoN). The new Ammonia Monitoring Network (AMoN) will provide another source of data to compare to the results of the model. The AMoN network is comprised of more than 60 sites (as of 2014) with passive samplers which provide 2-week average gaseous ammonia concentrations, often co-located with sites in the National Atmospheric Deposition Program sites.

1.6 Research Objectives

- Develop farm-level process-based models to track nitrogen through the production system for beef cattle, swine and poultry for farms throughout the United States and evaluate seasonal model performance based on data from published literature.
- Evaluate farm-level models using long-term observations from the National Air Emissions Monitoring Study (NAEMS) at seasonal and daily time resolution for swine, dairy, layer and broiler farms.
- Build a national inventory for ammonia emissions from beef, swine and poultry based on results from farm-level data, national animal population data (with county level resolution), and estimates of distributions of management practices and compare the inventory results to total livestock ammonia emissions in the 2011 NEIv2.

CHAPTER 2: SEMI-EMPIRICAL PROCESS-BASED MODELS FOR AMMONIA EMISSIONS FROM BEEF, SWINE, AND POULTRY OPERATIONS IN THE US

2.1 Abstract

Farm-level ammonia emissions factors in the literature vary by an order of magnitude due to manure management practices and meteorology, and it is essential to capture this variability in emission inventories used for atmospheric modeling. Loss of ammonia to the atmosphere is described here through a mass balance on nitrogen that incorporates a mass transfer resistance parameter, which varies with meteorological conditions and is tuned to match literature-reported emissions factors. The tuned parameters are specific to each set of management practices, while meteorological conditions may vary. Our farm emissions models (FEMs) explain between 20% and 70% of the variability in published emissions factors and typically estimate emission factors within a factor of 2. The R values are: 0.72 for swine housing (0.82 for shallow-pit houses); 0.48 for swine storage; 0.53 for broiler chickens; 0.87 for layer chickens; and 0.21 for beef feedlots (0.36 for beef feedlots with more farm-specific input data). Mean fractional error was found to be 22-44% for beef feedlots, swine housing, and layer housing; fractional errors were greater for swine lagoons (90%) and broiler housing (69%). Unexplained variability and errors result from: model limitations, measurement errors in reported emissions factors, and a lack of information about measurement conditions.

2.2 Introduction

Ammonia is a significant air pollutant because of its impacts to land, water, and human health through eutrophication, deposition, and the fine particulate matter formation (PM). Excessive ammonia leads to the eutrophication of pristine terrestrial ecosystems and many waterways (Draaijers et al., 1989). Deposition of nitrogen oxides and sulfur dioxide, has decreased in recent years; however, ammonia emissions have not decreased during this period (Driscoll et al., 2001; Fenn et al., 2003). Ammonia is a key component in the formation of fine PM (Ansari and Pandis, 1998), especially nitrate PM, as gas-phase nitric acid condenses only when ammonia is available for neutralization. Ammonium nitrate formation depends on: the amount of ammonia present, temperature, relative humidity and other pollutant concentrations (West et al., 1999). Because ammonium nitrate formation is temperature-dependent, the spatiotemporal variations in ammonia emissions can have a significant impact on the formation of PM. As ammonium nitrate formation is favored at colder temperatures, emissions in winter can have a greater impact on particulate matter formation than ammonia emitted in warmer seasons. The control of PM is so important because fine PM has been linked to respiratory ailments, cardiac events, and premature death (American Lung Association, 2006; Pope and Dockery, 2006).

Ammonia is emitted from several sources, but in the United States, the animal livestock sector contributes 70-90% of total national emissions (Battye et al., 1994, 2003; Bouwman et al., 1997; Pinder et al., 2006), mostly from dairy and beef cattle, swine, and poultry. Ammonia emissions from livestock are expected to increase as animal populations continue to grow and production intensifies to meet greater global demands (USEPA, 2004). Emissions of ammonia occur throughout the entire livestock production process—from animal housing, storages, and applied manure.

Early ammonia emission inventories relied on static emission factors (EF) to estimate the livestock contribution to the national ammonia emission inventory. However, it has been demonstrated that ammonia emissions are strongly dependent on meteorology, nutrition, and manure management; a single emission factor is unable to capture this variability (Ad Hoc Committee on Air Emissions from Animal Feeding Operation (National Research Council), 2003). The 2011 NEIv2 has added a temperature dependence to the emissions profile for ammonia, but the effects of management practices are still uncaptured. More recent work has attempted to account for some of this variability by using regression to relate an important parameter, e.g. temperature or animal age, to the emission factor (Rotz and Oenema, 2006), but it is still difficult to capture all the factors that cause variability in emissions.

Given the need to capture seasonal and regional variations in ammonia emissions for $PM_{2.5}$ modeling, the goal of this work is to build process-based models of ammonia emissions used for building national/regional emissions inventories. While complex, first principle process-based models may be able to characterize more emissions, they are computationally intensive and require detailed input data which is unlikely to be available for all farms and practices (Zhang et al., 2005). While they may reproduce emissions behaviors at specific farms with well-characterized conditions, their utility for building emissions inventories has not yet been demonstrated.

Our model is a balance between an empirical approach and first-principles process-based model. We use a nitrogen mass balance and a process description of ammonia losses, but tune model parameters to reproduce measured emissions factors. We limit model complexity to the most important emissions processes and to inputs that are typically available. The strategy pursued here for developing process-based models is guided by the need to build emissions inventories, and the requirements and data limitations associated with this application. Previous measurement campaigns also often sampled emissions from a single part of the production process. This means that we may not have information about the emissions process from the start to end of production, making nitrogen mass balance in the system difficult. The lack of whole-farm measurements is one gap in much of the literature available and a benefit of the estimates of ammonia emissions produced by the FEM.

In this chapter, we describe the adaptation and evaluation of previously developed process-based ammonia emissions models for beef cattle, swine, broiler and layer emissions based on the existing model framework called the FEM (for dairy cows) which conducts a mass balance on system nitrogen and water volume (Hutchings et al., 1996; Pinder et al., 2004a). Our model relies on input parameters to reproduce ammonia emissions for different farms, including meteorology, management practices and manure characteristics. The previous studies addressed only grazing cattle (Hutchings et al., 1996) and mature dairy cows (Pinder et al., 2004a).

2.3 Model Description

2.3.1 Model Overview

Our FEM captures ammonia emissions variability (caused by differences in meteorology, practices, and manure) through the use of a semi-empirical process-based model while constraining overall emissions via mass balance on available nitrogen in the farm system. For each livestock type, the FEM is composed of a series of submodels, each of which treats a different stage of manure management: housing (or grazing), storage, and application. The manure management trains for each livestock type are shown in Figures 2.1a-c, while the inputs, outputs, and time step of each submodel are shown in Figure 2.1d. Submodel configuration for each livestock type and management practices is detailed in Table 2.2.

The model uses inputs for farm type, manure nitrogen, and meteorological conditions to predict farm-specific ammonia emissions. Meteorological data are from the National Climate Data Center (NCDC), based on the time and location of the literature studies used in tuning if not directly provided. Farming practices and nitrogen inputs were based on literature-reported values for beef, swine and poultry from ammonia emissions measurement studies and animal nutrition research (Table 2.1).

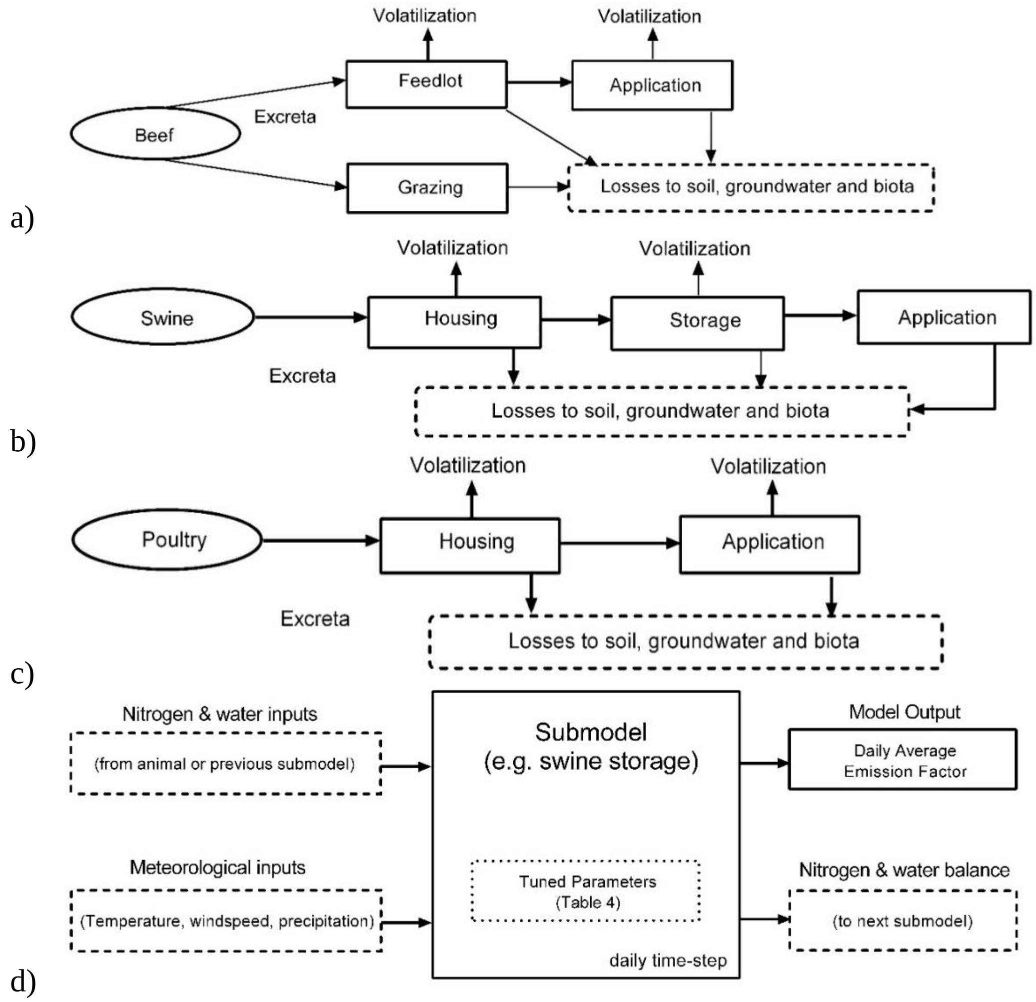

Figure 2.1. Waste and nitrogen flows used in the farm emissions models (FEMs) for a) beef cattle, b) swine, and c) poultry. Figure 2.1d shows how data flows through our submodels, showing how farm and meteorological input data are combined with the submodel's tuned parameters to produce emission factors and provide a mass balance which is passed along to subsequent submodels.

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Table 2.1. Key model inputs for beef, swine, and poultry manure management

Parameter Name	Animal Type	Range of Values	Value used *	Units	Source
Manure Volume	Beef cattle	12-17	15	liters $animal^{-1}$ day^{-1}	(Pinder et al., 2004b; VanHorn, 1998)
	Swine	4-10	6	liters $animal^{-1}$ day^{-1}	(Chastain et al., 1999)
	Poultry-Layer	0.088	0.088	liters $animal^{-1}$ day^{-1}	(Lacey et al., 2003)
	Poultry-Broiler	4.9	4.9	liters finished $animal^{-1}$	(ASAE, 2005)
Manure Urea Content	Beef cattle	47-70	60	kg N $animal^{-1}$ $year^{-1}$	(ASAE, 2005)
	Swine	11-30	19	kg N $animal^{-1}$ $year^{-1}$	(ASAE, 2005)
	Poultry-Layer	0.5-0.6	0.55	kg N $animal^{-1}$ $year^{-1}$	(ASAE, 2005)
	Poultry-Broiler	0.05-0.06	0.055	kg finished $animal^{-1}$	(ASAE, 2005)
Housing pH	Beef cattle	7.7	7.7		(Cole et al., 2009)
	Swine	6.5-7.5	7		(Lim et al., 2004)
	Poultry-Layer	7.1-7.6 (MB); 8.4-8.7	7.3		(Bolan et al., 2010; Liang et al., 2005)
	Poultry-Broiler	8	8		(Ferguson et al., 1998)
Storage pH	Swine	7.5 – 8	7.7		(Arogo et al., 2003)
Application pH	Beef cattle	7.5	7.5		(Rotz and Oenema, 2006)
	Swine	7.8 - 8.2	8		(Safley et al., 1992)
	Poultry-Layer	7.2	7.2		(Sommer & Hutchings, 2001)
	Poultry-Broiler	8.8	8.8		(Coufal et al., 2006)
Grazing pH	Beef cattle	7.7	7.7		(Pinder et al., 2004b)

Note: *default input values are used only when other data is unavailable.* MB=manure-belt; HR=high-rise layer housing

2.3.2 Animal Nitrogen

Nitrogen is used by animals for weight gain and growth, bodily maintenance, and commodity production, but animals do not use all the nitrogen that they are fed. Unused nitrogen is then excreted as waste. If we better understand nitrogen use efficiency, we can better constrain the amount of total ammoniacal nitrogen (TAN) available for volatilization as ammonia. Literature studies have shown that more waste nitrogen leads to higher manure ammonia emissions (Cole et al., 2006; Ferguson et al., 1998; Rotz, 2004; Todd et al., 2006).

Nearly of 80% of the nitrogen cattle are fed is excreted as urine or feces (Yan et al., 2007). For swine, 45-60% of dietary nitrogen intake is excreted as waste. Poultry can be divided into two types: broilers or layers, raised for their meat and eggs respectively. Broilers use 40-50% of their nitrogen intake, with the remainder excreted as waste (Jorgensen et al., 1990). For layers, fed nitrogen is partitioned between metabolism, egg production and waste, with 50-60% of dietary nitrogen excreted.

Animal wastes are composed of urine and feces. Most emissions come from urine, as its nitrogen is easily broken down into ammonia and the liquid allows for easier mass transfer than from feces. For beef, there are emissions from the solid fraction of wastes, but there is little information in the literature to constrain these emissions and so a constant emission factor of 3 kg yr^{-1} was used. For swine and layers, manure is stored as liquid slurry without a solid fraction that needs to be handled separately. For broilers, wastes are deposited on litter material in their housing and the waste is removed from the house after a production cycle and contains broilers' excrement and the litter material; manure characteristics are described (by type) in Table 2.1.

2.3.3 Ammonia Volatilization and Emissions Equation

The FEMs for each animal type, submodel, and practice have unique tuned parameter values, but the process is described by the same mass balance and mass transfer equations. All of the practices tuned in the model are described in Table 2.2 and the degree to which each submodel was tuned was dictated by the amount of literature data available. For "fully tuned" submodels, sufficient data was present in the literature to characterize the variability in emissions from that phase of production. For the submodels that were modified from the dairy FEM (Pinder et al., 2004b), less data was available in the literature, so the dairy submodel was used as a basis and adjusted. For beef application, where very little data was published, estimates of emissions were limited to constant emission factors of ammonia; in this case, we expect a small portion of the overall emissions to occur during this stage of manure management.

Table 2.2. Practices included in tuned farm submodels

Animal Type	Housing Submodel	Level of Tuning	Storage Submodel	Level of Tuning	Application Submodel	Level of Tuning	Grazing Submodel	Level of Tuning
Beef	Feedlot	Fully tuned	n/a	n/a	Solid/ broadcast application	Constant value used	Pasture	Based on modified dairy FEM
Swine	Deep pit, slatted floor	Fully tuned	Lagoon, Basin, Tank	Lagoon was fully tuned, basin and tank used modified dairy FEM	Injection, trailing hose, irrigation	Based on modified dairy FEM	n/a	n/a
Broiler	Litter system	Fully tuned	n/a	n/a	n/a	n/a	n/a	n/a
Layer	High-rise, manure belt	Fully tuned	n/a	n/a	n/a (transfer offsite)	n/a	n/a	n/a

Ammonia volatilization occurs at the surface of the liquid manure solution and is transported to the atmosphere above the wastes. Equation 1 is used as the basis for calculating emissions in each of the individual submodels of the beef, swine, and poultry models, previously used in the dairy FEM.

$$EF = A * [TAN] * H^{*} * r^{-1} \qquad (1)$$

$$r = r_a + r_b + r_s \tag{1b}$$

In this equation, the emissions factor (kg day^{-1} animal^{-1} or kg day^{-1} animal unit (AU) $^{-1}$) is described by *A*, the area fouled by excretion (m^2 animal^{-1}) that depends on the stocking density of the animals, often reported in the literature, *[TAN]*, the total ammoniacal nitrogen concentration in the waste (kg m^{-3}), the effective Henry's law constant, H^*, describing the equilibrium of the ammoniacal nitrogen in the system (dimensionless), and *r* is the mass transfer resistance (day m^{-1}). The resistance parameter, *r*, describes how mass transfer of ammonia between phases is inhibited. As in dry deposition models, this resistance consists of three components (Equation 1b): the aerodynamic (r_a), quasi-laminar (r_b), and surface resistances (r_s) (Wesely and Hicks, 1977). The aerodynamic and quasi-laminar resistances are used to describe the resistance to transport in the gaseous layer above the animal wastes (Olesen and Sommer, 1993; Sommer and Hutchings, 2001; Sommer and Olesen, 2000). The surface resistance is resistance due to diffusion through the layer of manure nearest the gas-liquid interface. Here, the surface resistance is tuned so modeled EFs agree with published literature data for the FEM.

Each submodel in the FEMs for beef cattle, swine and poultry rely on three mass balance equations that track the changes over time in manure volume, *V*, urea concentration and total ammoniacal nitrogen concentration (Equations 2-4).

$$\frac{dV}{dt} = k_{load} + k_p A - k_e A - k_i A \tag{2}$$

$$\frac{d[urea]}{dt} = k_{load}C_{urea}V^{-1} - k_{urea}[urea] - A[urea]k_iV^{-1} \quad (3)$$

$$\frac{d[TAN]}{dt} = k_{urea}C_T[urea] - A[TAN]H^*r^{-1} - A[TAN]k_iV^{-1} \quad (4)$$

The first equation describes how manure volume changes over time. Volume increases as manure is deposited to the surface (k_{load}) and by precipitation (for outdoor practices) as k_p (the precipitation loading rate), multiplied by the area where manure is deposited. Volume is lost via emission and infiltration, described by the k_eA and k_iA terms, respectively. Equation 3 describes how urea concentration changes over time; urea mass is added when waste is excreted and decreases via its hydrolysis, described by the $k_{urea}[urea]$term; infiltration below the surface is described by$A[urea]k_iV^{-1}$. Equation 4 describes how [*TAN*] changes in the system over time due to the increase as a result of urea breakdown, and losses due to volatilization and infiltration. The units of *k* depend on the equation, but each describes the rate at of a process (e.g. the units of k_{load} are L h^{-1}).

2.3.3.1 Housing Submodel

For beef, feces and urine are deposited to the feedlot surface. Thus, *d[TAN]/dt* is driven by temperature, wind speed, precipitation, and volume changes resulting from evaporation and infiltration. The r_s for beef feedlots is tuned as a function of temperature and wind speed, *u,* plus a constant resistance, *c,* of 3 s•m^{-1}.

$$r_s = H_1T + H_2u + c \quad (5)$$

Swine and poultry housing parameters are more similar in form to the earlier dairy FEM, where animals are housed enclosed and indoors (Equation 6). Thus, resistance can be estimated using only outdoor temperature and not wind-speed or precipitation. Outdoor temperature is used because it drives indoor ventilation rates and is correlated with emissions factors.

$$r_s = H_1 + H_2 T \qquad (6)$$

We have characterized two types of swine housing in our FEM: slatted floor (shallow-pit) and deep-pit housing. The first is found commonly in the Southeast and manure is removed frequently, leaving a high-nitrogen waste product in storage. Deep-pit housing is common in the Midwest and transfers manure less often, resulting in greater housing emissions and less in storage.

Poultry production includes two animal types: broilers and layers. Broilers are raised for meat and have a production cycle of approximately 6-7 weeks, while layers are raised for eggs and have a longer life cycle. Broilers reside in litter-based housing systems that are partially cleaned out after each cycle. Layers are housed in either high-rise or manure-belt houses. High-rise systems only remove manure every few months. In contrast, manure-belt systems remove manure frequently which limits housing ammonia emissions.

2.3.3.2 Storage Submodel

Storage emissions from the manure removed from beef feedlots have not been well-characterized in the literature so the beef FEM does not include a storage submodel.

Similarly, poultry manure storage receives little coverage in the literature so we are unable to create and tune a model for poultry manure storage. Swine manure storage, has been studied relatively extensively, and there are many ammonia EFs in the literature. In this submodel, we consider lagoons and storage basins. Resistance is related to the ability of the storage surface to form a crust. A crust inhibits mass transfer of ammonia from the wastes to the atmosphere.

2.3.3.3 Application Submodel

Manure application method is dependent on the manure dry matter content. Beef feedlot wastes are scraped as a solid from the surface of the feedlot after production. While emissions from the feedlot surface have been reported in the literature; emissions from the application of beef feedlot manure have not. As a result, we have used a constant emission factor of 3 kg year^{-1} in our submodel, which falls on the lower end of the range specified previously for dairy cows, since emissions from beef cattle are expected to be lower as a result of their lower dietary nitrogen requirements (Pinder et al., 2004b). Application emissions from swine have been better characterized in the literature. We have information about emissions from application via irrigation, trailing hose, and injection. Using data from the literature, we are able to tune model parameters to describe nitrogen infiltration. A_1 is a constant specific to application method, used in the calculation of the mass transfer resistance. Equation 7 then relates the infiltration rates used in Equations 2-4 to soil permeability, *Ki*, and *DMC*, dry matter content to the rate of infiltration, which affects ammonia volatilization. The infiltration rate is further constrained by parameters A_2 and A_3, which are unitless, tuning to ensure agreement with literature-reported EFs.

$$k_i = K_i \times 10^{[A_2 + A_3 DMC]} \quad (7)$$

Too little data exists in the literature to constrain estimates for emissions from the application of poultry manure for broilers or layers. Additionally, a significant fraction of poultry manure is re-processed for use in feed and other applications.

2.3.3.4 Grazing Submodel

Emissions from beef cattle raised on pasture were treated similarly to dairy cattle raised on pasture, increasing the excreted nitrogen because they are not producing milk. Additionally, using the limited literature data available, we made sure our submodel produced reasonable estimates of ammonia emissions. This submodel has previously been described in work on mature dairy cattle by Pinder (2004). Emission factors for grazing cattle in the literature range from <5 g NH_3/animal/day (Van Der Hoek, 1998) to greater than 30 g NH_3/animal/day. The FEM typically predicts emission factors of 5-20 g NH_3/animal/day.

2.3.4 Ammonia Equilibrium and Nitrogen and Volume Changes

Ammonia emissions from livestock result primarily from the breakdown of urea in the animals' urine by the urease enzyme (Udert et al., 2003) into ammonia, which partitions into the air and waste as ammonia gas and ammonium in solution. Our model assumes a 2-hour timescale (Monteny and Erisman, 1998) for the breakdown of urea into TAN and proceeds once waste nitrogen has been converted into TAN. In manure liquid, ammonium and ammonia reach equilibrium according to their acid-base equilibrium constant, K_a. After equilibrium is reached, ammonia is partitioned between the waste's aqueous phase and the gas phase above it. The partitioning can be described by Henry's law constant, K_h. The effective Henry's law constant can then be described as a ratio of the gas phase ammonia to the total ammoniacal nitrogen in the solution, H^*, and described in previous work by Pinder, et al., shown in Equation 8:

$$H^* = \frac{[NH_{3(g)}]}{[TAN]} = \frac{K_a}{K_h[H^+] + K_a(1 + {K_h}^{-1})} \qquad (8)$$

TAN concentration depends on the amount of nitrogen in the excreta and the volume of excreta. A more concentrated sample will have a higher *[TAN]*, so processes affecting manure volume impact equilibrium. Volume can be affected by: precipitation (increasing volume), soil infiltration, and runoff (decreasing volume). The submodels incorporate both precipitation and infiltration into the characterization of ammonia emissions from beef, swine, and poultry. Runoff is not explicitly treated because significant runoff is largely coincident with low ammonia emissions and would not contribute significantly to the overall ammonia emissions profile (Todd et al., 2008).

2.3.5 Input Data and Parameter Tuning

Input data were taken from the literature to better constrain the model to reported farm conditions (Table 2.1). Values used are selected because they fell in the middle of the range reported in literature. Several papers reported manure nitrogen content for a range of fed nitrogen values, as well as the emissions of ammonia from these animals, so specific values were used (where available) because ammonia emissions are highly sensitive to this parameter.

To ensure our FEMs produce realistic EFs, we used literature data to tune parameters related to r_s in mass transfer equations (Equations 2-4). Their descriptions and values are in Tables 2.3-2.4.

Table 2.3. Tuned model parameters for beef, swine, and poultry

Submodel	Animal Type	Description	Tuning/Evaluation Sources
Housing	Beef cattle, Swine, Poultry-Layer, Poultry-Broiler	Resistance parameters H_1, H_2 (Eq. 6)	1- 4
Storage	Swine	Resistance parameters S_1, S_2	5
Application	Beef cattle, Swine, Poultry-Layer, Poultry-Broiler	Resistance parameters A_1,A_2, A_3 (Eq. 8)	6-9
Grazing	Beef Cattle	Resistance parameters G_1, G_2 (similar to Eq. 8)	10

1. (Cole et al., 2006; Hristov et al., 2011; Klopfenstein and Erickson, 2002; Todd et al., 2011, 2008, 2007, 2006, 2005)
2. (Aarnink et al., 1995; Arogo et al., 2003; Heber et al., 2000; Hoff et al., 2006; Jacobson et al., 2005)
3. (Fabbri et al., 2007; Liang et al., 2005; Nahm, 2003; Nicholson et al., 2004)
4. (Casey et al., 2003; Coufal et al., 2006; Gates et al., 2008; Lacey et al., 2003)
5. (Harper and Sharpe, 1998; Lim et al., 2003; Osada et al., 2000; Portejoie et al., 2003; Visscher et al., 2002; Zahn et al., 2000)
6. (James, 2008; McGinn and Sommer, 2007)
7. (Chantigny et al., 2007; Sharpe and Harper, 1997; Westerman et al., 1995)
8. (Pelletier, 2008; Redwine et al., 2002)
9. (Sommer and Hutchings, 2001)
10. (Hatch et al., 1990)

Table 2.4. Practice-specific parameter values for beef, swine, and poultry

Submodel	Animal Type	Description	Parameter Values
Housing	Beef cattle, Swine, Poultry-Layer, Poultry-Broiler	Beef Feedlot	$H,=0.1\ (s \bullet m^{-1} \bullet {}^{\circ}C^{-1}),\ H_2=-0.01\ (s^2 m^{-2})$
		Swine—shallow pit	$H,=0.08(s \bullet m^{-1}),\ H_2=-0.004(s \bullet m^{-1} \bullet {}^{\circ}C^{-1})$
		Swine—deep pit	$H,=0.1(s \bullet m^{-1}),\ H_2=-0.008(s \bullet m^{-1} \bullet {}^{\circ}C^{-1})$
		Layer—Manure belt	$H,=0.3(s \bullet m^{-1}),\ H_2=-0.015(s \bullet m^{-1} \bullet {}^{\circ}C^{-1})$
		Layer—High Rise	$H,=0.22(s \bullet m^{-1}),\ H_2=-0.02(s \bullet m^{-1} \bullet {}^{\circ}C^{-1})$
		Broiler	$H,=0.15(s \bullet m^{-1}),\ H_2=-0.035(s \bullet m^{-1} \bullet {}^{\circ}C^{-1})$
Storage	Swine	Swine lagoon	$S_1=0.20(s \bullet m^{-1}),\ S_2=4.00(s \bullet m^{-1} \bullet {}^{\circ}C^{-1})$
		Swine basin	$S_1=0.11(s \bullet m^{-1}),\ S_2=2.24(s \bullet m^{-1} \bullet {}^{\circ}C^{-1})$
Application	Beef cattle, Swine,	Beef—broadcast	$A,=0.0004,\ (s \bullet m^{-1}) A_2=0.88,\ A_3=-1.4$
		Swine—irrigation	$A,=0.001(s \bullet m^{-1}),\ A_2=-10,\ A_3=20$
		Swine—injection	$A,=0.01(s \bullet m^{-1}),\ A_2=-15,\ A_3=40$
Grazing	Beef Cattle	Beef Pasture	$G,=0.12(s \bullet m^{-1}),\ G_2=5.4$

The model's tuned parameters are set for each practice, and these tuned parameters have been constrained by the emissions factors that have been reported in the literature for those specific housing, storage, and application practices (across a variety of seasons and locations). The FEM could then be run using these tuned parameters for different meteorological conditions in order to produce estimates for a farm in a different location. Previous work on dairy cows by Pinder (2004) used 2 studies to tune the housing submodel, 3 for storage, 4 for the application submodel, and 3 for grazing, or 12 studies total to tune the dairy submodels. We used 6 studies to tune the beef feedlot submodel, 4 studies to tune the swine housing submodel and 6 for swine storage and 2 studies were used for broiler and layer housing each, details in Table 2.5 below.

Table 2.5. Data sources used in model tuning/evaluation for beef, swine, and poultry

Animal	Submodel	Data sources for tuning	Data sources for evaluation
Beef	Housing (feedlot)	1	1, 2
	Application	11	11
Swine	Housing	3	3, 4
	Storage	9	9, 10
	Application	12	12
Broilers	Housing	5	5, 6
	Application	13	13
Layers	Housing	7	7, 8
	Application	14	14

1. (Cole and Defoor, 2006; Hristov et al., 2011; Todd et al., 2008, 2007, 2006, 2005)
2. (Klopfenstein and Erickson, 2002; Todd et al., 2011)
3. (Aarnink et al., 1995; Heber et al., 2000; Hoff et al., 2006; Jacobson and Hetchler, 2005)
4. (Arogo et al., 2003)
5. (Burns et al., 2007; Lacey et al., 2003)
6. (Casey et al., 2003; Coufal and Chavez, 2006)
7. (Fabbri et al., 2007; Nicholson et al., 2004)
8. (Liang et al., 2005; Nahm, 2003)
9. (Harper and Sharpe, 1998; Lim et al., 2003; Osada et al., 2000; Portejoie et al., 2003; Visscher et al., 2002; Zahn et al., 2000)
10. (Arogo et al., 2003)
11. (James, 2008; McGinn and Sommer, 2007)
12. (Chantigny et al., 2007; Sharpe and Harper, 1997; Westerman et al., 1995)
13. (Pelletier, 2008; Redwine et al., 2002)
14. (Sommer and Hutchings, 2001)

2.4 Results

Ammonia emissions depend strongly on temperature and nitrogen intake and less on wind speed and precipitation. This is reflected in our emission estimates from the FEMs. However, literature-reported emission factors often fail to report key contextual information that could be used to better constrain the farm emission model input parameters. Additionally, emissions from some practices have been left underreported or unreported.

The predictions of FEMs for beef cattle, swine, and broiler and layer chickens are presented and evaluated here. Ammonia emissions are generally predicted to have a strong seasonal cycle. There are regional trends in emissions—driven by the temperature dependence of ammonia volatilization and the management practices used. Single variables aren't able to capture fully the variability that has been observed in literature EFs, so our model uses a number of readily available inputs for simulation. Data from the literature are provided at various timescales, from a few days, to weeks or seasonal average emissions. For consistency, we have reported our model evaluation in terms of *g* NH_3*/animal/day* or *g* NH_3*/animal unit/day*.

2.4.1 Model Evaluation

Results from the submodels of the FEMs are described in this section. The *R* value (Figure 2.2a) for beef feedlots is 0.45. Figure 2.2b shows the evaluation for beef feedlots where information about feed/excreta nitrogen content is known, which has an *R* value of 0.62. Figure 2.2c shows the swine housing evaluation with an *R* value of 0.67 (0.81 for shallow-pit only); Figure 2.2d shows the swine storage results with an *R* value of 0.69; lower observed EFs often come from flux chamber measurements while higher emissions are observed from inverse dispersion measurements in which downwind concentrations are measured and the emissions are calculated through the use of a dispersion model. Poultry EFs are presented as grams of ammonia volatilized per animal unit (AU) per day, where 1 AU is 500 kg, which separates the effects of animal growth from other variability. Broilers show an *R* value of 0.53 and layer housing had an *R* value of 0.87 (with a clear separation between the two major housing types used). Emission factors recommendations from earlier EPA guidelines fall within the range of emissions factors produced by the FEM (Faulkner and Shaw, 2008; USEPA, 2004).

Ammonia is difficult to measure resulting in significant uncertainty in the literature. Our modeled EFs also have substantial uncertainties because we used a simple model for emissions, using a few readily available parameters, rather than using first-principles-based model with more detailed inputs. Specifically, passive sampling devices are operated with a goal of less than 30% uncertainty (range of uncertainty is 15-80%) (Kirchner et al., 1999) while emissions estimates from the use of backwards Lagrangian stochastic models is estimated to be 15-20% or greater for unstable conditions (Flesch et al., 2004). A summary of the correlations, mean fractional error (MFE), normalized mean error (NME), and mean fractional bias (MFB) is in Table 2.6.

Table 2.6. Model evaluation statistics for beef, swine, and poultry FEMs

Animal Type	Model vs. Measured *R*	Mean Fractional Error	Normalized Mean Error	Mean Fractional Bias
Beef Feedlots	0.45	44%	38%	+4.4%
Beef Feedlots (Adjusted)	0.62	31%	29%	+1.7%
Swine Housing	0.72 (all)	37%	28%	-8.7%
Swine Lagoon	0.69	90%	61%	+11.9
Broiler Housing	0.53	69%	55%	+1.7%
Layer Housing	0.87 (all) 0.39 (man. belt) 0.67 (hi-rise)	22%	24%	-2.5%

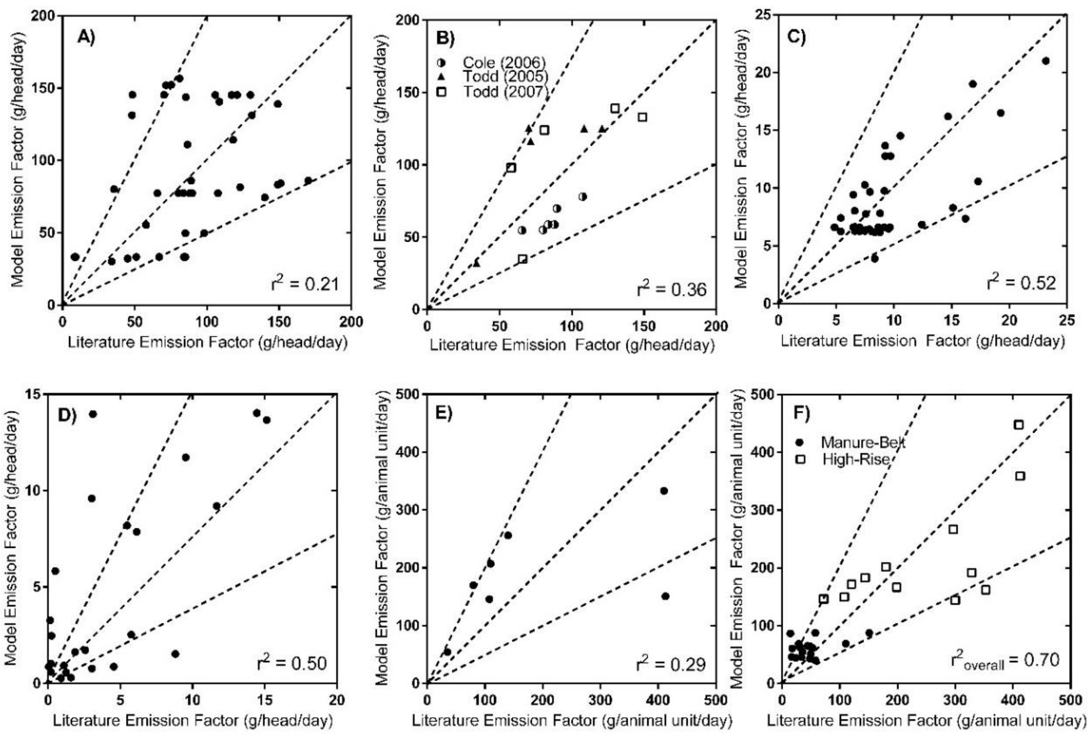


Figure 2.2. Model evaluation for: a) beef feedlots evaluated against all EFs; b) beef feedlots where nutritional inputs and/or nitrogen excretion is known; c) swine housing; d) swine storage lagoons; e) broiler housing; f) layer housing emission factors evaluated against literature data for high-rise (HR) and manure-belt (MB) barns.

Measuring open sources like feedlots often relies on a two-step monitoring technique that includes measurement and modeling components; thus there is greater uncertainty in these measurements and greater scatter in their emission factors. Comparing the NME for each of the submodels shown in Figure 2.2, the values for beef feedlots, swine, and layer houses of 25%-60% are comparable in magnitude to the uncertainties in the measurement techniques used by the studies in literature. Though our model may omit or simplify important processes contributing to emissions variability, we conclude that a significant fraction of the model-measurement discrepancy may reflect limitations of the measurements. Given that we tune our models to the literature, normalized mean bias in our model predictions is small.

2.4.2 Trends in Modelled Ammonia Emissions

Literature-based ammonia emissions factors are highly variable, particularly for open sources. These measurements are difficult to conduct with great enough time resolution to capture brief spikes and dips in emissions. Additionally, beef feedlots, swine houses and swine lagoons ammonia emissions exhibit a strong seasonal pattern.

For beef feedlots, the value of better contextual information, specifically feed and manure nitrogen content, in producing accurate estimates of ammonia emission factors is highlighted in Figures 2.2a and 2.2b. The model performs better with location-specific information for inputs, with R value improving from 0.45 to 0.62 and reducing NME from 38% to 29%. Many EF measurements fail to report contextual information, particularly manure nitrogen content, pH, and meteorological conditions, which would enable better interpretation and comparison of measurements, understanding of variability, and better emission models and inventories. Future emissions measurements should document better these conditions.

Feedlot emissions are much higher than pasture ammonia emissions. In this work, the grazing model was adapted from previous work (Pinder et al., 2004a), tuned with dairy and beef cattle data because of limited data. Since cattle on pasture comprise more than 80% of the total population in the United States at any given time (USDA-NASS, 2015), though their emission factors are 5-8 times lower than those on feedlots, we expect they contribute significantly to national emissions, and so measurements of pasture emissions should be a priority.

Figure 2.3 presents emissions at typical farms in Central Iowa and Eastern North Carolina, which were selected because of their importance in hog production (NPPC, 2012). By comparing practices and locations, we are able to identify the effects of location and practices on ammonia emissions. We see that having lower emissions in one stage of production means that there may be more nitrogen available for volatilization later on, altering when emissions happen, but not necessarily the amount. Comparing Figure 2.3b to 2.3d, we see the effects that different meteorological conditions have on ammonia emissions, with North Carolina, having nearly 1.5 times the emissions predicted in the cooler state of Iowa.

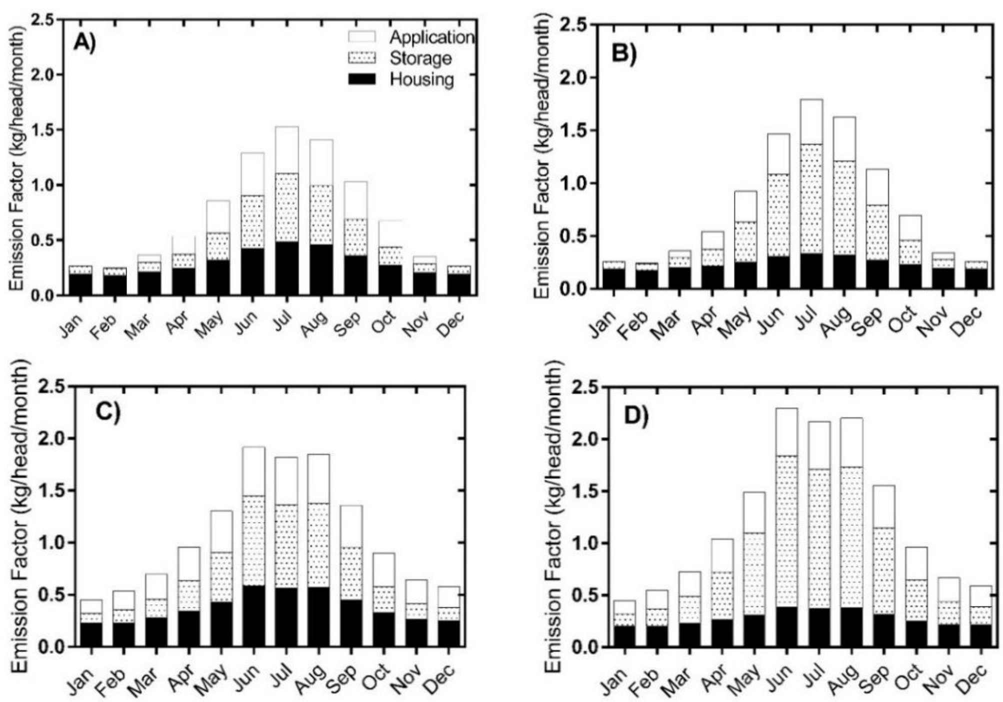


Figure 2.3. Sample swine farms in Iowa (IA) and North Carolina (NC): a) IA farm with deep-pit housing, basin storage, irrigation application; b) IA farm with slatted-floor, lagoon, irrigation application; c) NC farm with deep-pit housing, basin storage, irrigation application; d) NC farm with slatted-floor housing, lagoon storage, irrigation application.

Broiler chickens and layer chickens have different lifecycles. For broilers, temperature is not a driver of ammonia emissions because of the varying animal feed nitrogen contents during the production cycle, the animal growth rate and the animal nitrogen use efficiency. As a result, the FEM performs with only an R value of 0.53, suggesting that factors not used in our model (not typically available from measurement campaigns) contribute significantly to variability. Additionally, there is significant variability within measurements taken under similar temperatures and bird ages in the literature, shown in the range of literature emission factors from 30 g/AU/d to more than 400 g $AU^{-1}d^{-1}$, while the FEM ranges from 20-300 g $AU^{-1}d^{-1}$ (Figure 22.e).

Layer chickens are primarily housed in two kinds of barns: manure-belt and high-rise systems—with different sets of housing parameters in the FEM accounting for major differences in practices. Manure-belt systems are characterized by daily to weekly waste removal while high-rise houses remove manure less frequently. The time the manure spends in housing helps explain the differences in ammonia EFs between housing types for similar nitrogen inputs and animal characteristics (Figure 2f). The observation of higher emissions from high-rise barns is well-captured by the FEM, consistent with literature measurement campaigns. Temporal variability in ammonia emission factors is better captured for layers than broilers because animals maintain the same size, diet and environmental requirements during their time in the barn, reflected in the greater R value (0.87) for layers than broilers. The ability of the layer FEM to distinguish between housing types is important as both are used in production where 70% of layer facilities are high-rise configurations while 30% of facilities use manure-belt houses (Xin et al., 2011).

There is a lack of data in the literature regarding the long-term storage and endpoint of manure for poultry, whether it is recycled or land-applied or used in feed production. Due to limited data, we are unable to well-characterize the spatiotemporal variability in ammonia EFs for these parts of manure management. However, we expect housing is the most significant source of ammonia emissions for poultry, so this does not appear to be a serious limitation.

2.5 Conclusions

Ammonia emissions from livestock operations are highly variable and depend on manure management practices and meteorological conditions. Representing this variability in emissions inventories is challenging but essential, and process-based modeling offers one promising approach. In this work, farm emissions models (FEMs) were developed for the most common livestock production practices for beef cattle, swine, and broiler and layer chickens. Building on previous work (Pinder et al., 2004a; Hutchings et al., 1996) for dairy cows, the FEMs are based on mass balances of the nitrogen and water flowing through the farm using model inputs from literature and tuned model parameters for livestock in the United States. Key inputs to our FEMs include manure nitrogen contents, manure volumes per animal, and manure pH throughout production. This method of using literature-based inputs and a constant set of tuned parameters (for each set of practices) means our model is semi-empirical, but captures much of the variability lacking from earlier emissions inventories. Moreover, these FEMs are relatively simple and computationally efficient to run, meaning they can be used to build national emissions inventories given meteorological inputs, animal populations, and management practices.

Model performance was evaluated in terms of the *R* value between literature and modeled EFs, mean fractional error, and mean fractional bias (based on the difference from observed emissions factor) (Table 2.6). The *R* values show that the FEMs capture 20%-70% of the variability that is seen in literature emissions factors. For swine, the housing *R* value was 0.72 and 0.69 for lagoon storage; *R* was 0.53 for broilers and 0.87 for layers; for beef feedlots R values were 0.45 for all feedlots and 0.62 for feedlots with farm-specific nitrogen information. FEM performance was better for enclosed emission sources than open sources. Open-air production strategies produce greater ranges in measured emission factors and cause greater uncertainty in our model-predicted values due to the measurement techniques which require concentration measurement and dispersion modeling to produce an estimate of ammonia emissions (Flesch et al., 2007).

We found that model performance could be improved with better input data about feed and manure nitrogen contents, which are not always reported in the literature. Our FEM was more successful in predicting reported emissions factors for cases where feed and/or nitrogen contents were reported (R value of 0.36) versus cases where they were not reported (R value of 0.21). Mean fractional error was also reduced (from 44% to 31%). This underlines the importance of reporting the conditions under which emissions are measured, including weather conditions, details of management practices, feed and manure nitrogen contents, and manure pH. Having this information accompany emissions factor measurements greatly increases the value of the emissions measurements especially for emissions model development. If more farms provided more details about manure nitrogen content, model performance could be improved substantially.

Model performance was limited by the lack of data in the literature for some common management practices for livestock production. This results from a tendency to focus on concentrated, intensive operations (important for nuisance and odor issues) to the exclusion of widespread sources that may be significant for national inventories, evidenced by a lack of literature data for pasture-raised beef though they likely contribute significantly to the overall national emissions. Additionally, there was a lack of data for the storage and application of poultry manure in the literature.

The next chapter evaluates the the dairy, swine and poultry FEMs with data from the National Air Emissions Monitoring Study (NAEMS), a more than two-year measurement campaign completed at a number of farms in several US states. This allows us to improve the process-based farm models developed by providing more highly-resolved and complete input information. It also highlights some of the regional differences in production methods that are important in understanding emissions during the production process.

2.6 Disclaimer

We have developed a farm-level model to better capture the major drivers of seasonal and regional variability in ammonia emissions from livestock production, but individual farms may differ significantly from these predicted due to farm-specific practices not represented here.

CHAPTER 3: EVALUATING A PROCESS-BASED MODEL FOR LIVESTOCK AMMONIA EMISSIONS USING LONG-TERM MEASUREMENTS

3.1 Abstract

Ammonia emissions from agriculture, especially livestock, are difficult to characterize due to the many sources of emissions variability (meteorology, management practices, and nutritional inputs) as well as difficulties in measuring emissions, especially from outdoor sources. Previous evaluation and tuning of our process-based farm emission model (FEM) was based on short-term measurements from the literature; given long-term emissions measurements provided by the National Air Emissions Monitoring Study, here we present additional evaluation and improvement of this model. The National Air Emissions Monitoring Study (NAEMS) is a recent measurement campaign encompassing a number of large-scale farms with dairy cows, swine, broilers and layers. These farms are located in a variety of states, demonstrating regional trends in production practices and allowing us to understand more about not only differences driven by meteorology but also by other sources of farm-to-farm variability. Emissions were measured at most farm across all seasons (for lagoons) and consistently for at least a year for animal houses, providing unprecedented information about the seasonal cycle of emissions as well as emissions under colder temperatures. We investigated the model's performance in predicting seasonal and daily variability in ammonia emissions from the NAEMS farms for dairy, swine, layers and broilers. Average seasonal R values were 0.69 for dairy housing and 0.8 for dairy storage, 0.49 for swine housing and 0.81 for swine storage, 0.87 (excluding farm NC2B) and 0.54 for all farms for layer

housing and 0.56 for broiler housing. Except for one dairy farm in California, mean fractional errors in housing were less than 61% with a median value of 22%. Storage emissions showed a greater range in error and biases (22 to 91% and -90 to 94%, respectively) due to the limited nature of our model and difficulties associated with measure open-source ammonia emissions. Daily evaluation of the FEM produced more mixed results with 9 farms having an R-value greater than 0.25, 5 farms with little to no correlation (R= -0.04 to 0.23), and 1 farm with an R-value of -0.76. Additionally, the NAEMS observations for layer chicken barns show a positive correlation between emissions and temperature on a daily time scale but a negative correlation on a seasonal time scale, possibly explained by the rate at which manure dries. Overall, this analysis demonstrates that the process-based FEMs perform reasonably well in predicting the magnitude of ammonia emissions, their seasonal cycle, and farm-to-farm variability and have some skill in predicting daily variation in emissions.

3.2 Introduction

Ammonia is an air pollutant of significant concern because it can enhance the formation of fine particulate matter (PM) as well as cause damage via deposition to land and water. Fine PM or $PM_{2.5}$ (the mass concentration of particles with aerodynamic diameters of 2.5 μm or less) has been shown to cause respiratory ailments, cardiac events, and premature death (American Lung Association, 2006; Krewski et al., 2003; Pope and Dockery, 2006). As a result, the control of PM has been shown to offer significant social benefits and has therefore been pursued aggressively (American Chemical Society, 2011; Fann et al., 2009). Beyond its health impacts (as a major component of particulate matter), excess ammonia deposition can lead to the eutrophication of pristine terrestrial ecosystems and many waterways (Erisman et al., 1998; Jenkinson, 2001). Ammonia emissions have been increasing, and are expected to continue to increase, as demand for animal products has grown and the livestock industry has been intensified. Some sources estimate an increase of ammonia emissions of 15% by 2030, whereas emissions of sulfur dioxide and nitrogen oxides, the other major precursors to inorganic fine PM, have decreased by 80% and 60% respectively in the past 30 years (Driscoll et al., 2003; Fenn et al., 2003; USEPA, 2004).

The major source of ammonia emissions in the United States is livestock, with the vast majority of livestock emissions resulting from beef, dairy, swine and poultry production (Battye et al., 1994, 2003; Bouwman et al., 1997; Goebes et al., 2003; USEPA, 2004; Webb and Misselbrook, 2004). Because both manure management practices and environmental conditions cause variability in ammonia emissions, not all management practices and meteorological conditions can be measured. Many times, measurement campaigns sample emissions only for a brief period or for a particular part of the production process. Each stage of the production process has associated emissions and though it is vital to know what is happening at each stage of manure management, often, we do not have information about the emissions process from the start to end of production which makes it difficult to complete a mass balance on the nitrogen in system. The lack of whole-farm measurements is one gap in much of the literature available and one aspect of ammonia emissions our development of a farm emission model attempts to remedy.

Previously, we developed a farm emissions model (FEM) some of whose parameters were tuned to match measurements reported in the literature for beef, swine and poultry production (McQuilling and Adams, 2015). Additionally, we have used the FEM developed previously by Pinder et al. for dairy production in the United States (Pinder et al., 2004a, 2004b). The farm emission model (FEM) developed is a process-based model that tracks the flow of total ammoniacal nitrogen and manure volume through the farm system by conducting a mass or volume balance through the whole system, ensuring mass is conserved. Although these process-based FEMs have been useful, significant limitations have remained, stemming from the lack of long-term, detailed observations with which to evaluate the model's performance in capturing seasonal variability. Understanding seasonal variability in emissions is vital because the role that ammonia plays (in terms of $PM_{2.5}$ mass) is typically much greater in the winter than summer (Ansari and Pandis, 1998; Stanier et al., 2012).

The National Air Emissions Monitoring Study came about as the result of the Animal Feeding Operations Consent Agreement and Final Order (AFO-CAFO) to better understand the effects of intensive animal production on the environment, and in particular, the quantity of atmospheric emissions of ammonia, hydrogen sulfide, non-methane VOCs, and particulate matter (Key et al., 2011; Mittal, 2009; Thompson, 2011). In this work, we use data from the NAEMS to fill in the gaps from more traditional measurement campaigns. The farm-specific feed and waste composition data, daily meteorological observations, and the length of observation (multiple years) were especially beneficial in this regard. In particular, the multi-year observations allowed us to better characterize seasonal variability in emissions while the data about manure characteristics allowed us to use more farm-specific data as model inputs. The daily wintertime observations were of special interest because a wintertime spike in ammonia emissions could cause high levels of PM formation, as observed especially in the Midwestern US, and so a single day could cause a particular location to be out of attainment for the particulate matter National Ambient Air Quality Standard (Stanier et al., 2012).

In this chapter, we describe the evaluation of our FEM against the data collected during NAEMS for housing emissions of ammonia from dairy cattle (using the tuned FEM from Pinder and Pekney, 2004), swine, broiler chickens, and layer chickens as well as for storage emissions from swine and dairy production at a variety of farm locations and practices described in more detail in the following sections. The model performance for the NAEMS data was reviewed in three ways. First, emissions factors from the prior literature were compared to the observations made during NAEMS. Because there were substantial differences in both the emissions factors as well as the conditions under which the observations were made, the FEM tuned mass transfer resistance parameters were re-tuned to account for both prior literature and newer NAEMS measurements. Then, the model was evaluated for its ability to reproduce the seasonal emissions patterns observed on the NAEMS farms; mean fractional error and mean fraction bias are reported as well as the correlation between model and observations. Additionally, the daily predicted emission factors were compared to the daily emission factors reported by NAEMS. Finally, we characterize the annual average performance of the model, separated by both animal type and specific farm practice and management stage.

3.3 Method Overview

3.3.1 Description of Farm Emission Model (FEM)

As previously described (Hutchings et al., 1996; McQuilling and Adams, 2015; Pinder et al., 2004a), ammonia emissions from livestock are driven by three major factors: meteorological conditions (e.g. temperature, wind speed and precipitation), manure management practices (housing type, storage type, fed nitrogen), and manure characteristics (volume, waste nitrogen content, etc.). The FEMs evaluated here were developed for the purpose of building national emissions inventories, which requires capturing the seasonal variability in ammonia emissions (driven in many cases by seasonal differences in temperature) as well as regional differences in emission patterns resulting from regional differences in farming practices. The submodels in the FEMs for dairy cattle, swine and poultry rely on three mass balance equations to characterize the changes over time in manure volume, urea concentration and total ammoniacal nitrogen concentration. More specifically, the FEM is a semi-empirical process-based model that uses a mass balance on nitrogen through the system to characterize the emissions process through the entire farm system (from housing to storage to manure application); it is semi-empirical because mass transfer model parameters are tuned to ensure output agrees with measured emission factors. For each livestock type, the farm emission model (FEM) is composed of a series of submodels, each of which treats a different stage of manure management: housing (or grazing), storage, and application. Configuration of the sub-models differs for each of the livestock types and management practices used. A schematic of the farm emission model is shown in Figure 3.1. In general, total ammoniacal nitrogen (TAN) and liquid volume are tracked as manure move from housing to storage to application. Where appropriate, e.g. dairy and beef cattle, a grazing sub-model is included as well. Storage of poultry manure and manure application was not included in this evaluation because those processes were not observed in NAEMS.

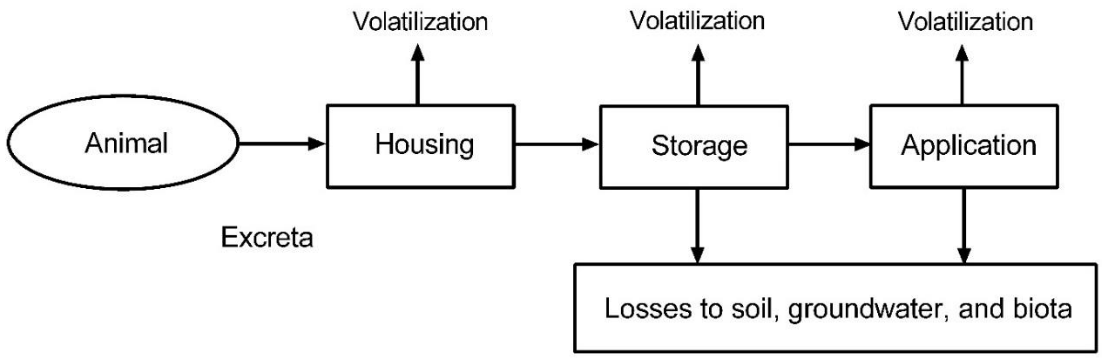


Figure 3.1: Schematic of farm emission model (FEM) previously described (McQuilling and Adams, 2015; Pinder et al., 2004a, 2004b).

Within the FEMs, we use inputs for management practices, manure nitrogen, and meteorological conditions to predict ammonia emissions from a particular farm type in a particular location, based on the data reported for that farm. A description of model inputs and parameters and their sources is shown in Table 3.1. Meteorological data were collected simultaneously with emissions measurements during NAEMS, and so we have detailed information about temperature, wind speed, and wind direction for each of the farms throughout the study. Additionally, manure and feed characteristics were also observed at various points during the study at the NAEMS farms (every 3-6 months), and data including the nitrogen content of manure, the manure pH, as well as feed nitrogen content were reported throughout the measurement campaign at a similar frequency and is used as input to our FEMs. As previously discussed, the level of detail in the meteorological observations and manure conditions helped to constrain the results of the FEM, as these data were often unreported in previous work.

Table 3.1: Description and sources of model inputs and parameters

Data Type	Description	Source of input or parameter	Input or Tuned Parameter?
Meteorology	Temperature (°C) Wind speed (m/s) Precipitation	NAEMS; unique for each farm NAEMS; unique for each farm From National Climate Data Center, based on farm location	Input value (monthly average for seasonal evaluation, daily for daily evaluation)
Manure Management Practice	Type of housing or storage	NAEMS; unique for each farm	Input value
Resistance Parameters	Surface mass transfer resistance from manure to atmosphere	Tuned based on literature and NAEMS observations to agree with previous work; constant for a particular management practice	Tuned Parameters

In general, the year-round NAEMS study provides many more measurements at colder temperatures for most animals and practices and a number of high emissions measurements for broiler housing and swine storage. Given these qualitative differences between the NAEMS data and prior literature, we decided to re-tune the FEM data against a combination of NAEMS and prior literature measurements. Therefore, the results here evaluate the ability of the FEMs to capture variability in ammonia emissions rather than a fully independent prediction of emissions.

In the FEM, as described previously (Hutchings et al., 1996; McQuilling and Adams, 2015; Pinder et al., 2004a), ammonia emissions are estimated as a function of the nitrogen present in the waste and the mass transfer resistance. This resistance is the made up of the following three parts: the aerodynamic (r_a), quasi-laminar (r_b), and surface resistances (r_s) (Wesely and Hicks, 1977). Aerodynamic and quasi-laminar resistances are used to describe the resistance to transport in the gaseous layer above the animal wastes (Hutchings et al., 1996; Olesen and Sommer, 1993; Sommer and Hutchings, 2001). These parameters are based on widely used theoretical formulas and are not tuned. The third part of the resistance is the surface resistance from diffusion closest to the gas-liquid (manure) interface. Here, the surface resistance is a function of tuned parameters as well as temperature which ensures the modeled ammonia emission factors are consistent with observations.

3.3.2 Description of National Air Emissions Monitoring Study (NAEMS) Data

The National Air Emissions Monitoring Study was a measurement campaign aimed at better exploring the seasonal and regional differences in emissions from livestock production in the United States. Farms were selected to span a range in practices as well as locations. The study was conducted from 2007-2010 and data was released to the public in 2011. Descriptions of the farm locations, animal types, manure management practices, and length of study are listed in Table 3.2 (USEPA-OAQPS, 2015b).

Table 3.2: Description of farms in the National Air Emissions Monitoring Study (NAEMS) including practices used, animal type and observation dates.

Farm Name	Animal Type	Management Stage	Practice Used	Dates of Study
CA1B	Broiler	Housing	Litter-based	December 2007 – October 2009
KY1B			Litter-based	February 2006 – March 2007
CA2B	Layer	Housing	High-rise	October 2007 – October 2009
IN2H			High-rise	June 2007 – May 2009
NC2B			High-rise	January 2008 – December 2009
IN2B			Manure-belt	January 2008 – December 2009
IA4B	Swine	Housing	Shallow pit/Flush (<24 d)	July 2007 – September 2009
IN3B			Deep pit (180 d)	July 2007 – July 2009
NC3B			Shallow pit/Flush (7 d)	December 2007 – December 2009
NC4B			Shallow pit/Flush (7 d)	December 2007 – December 2009
OK4B			Shallow pit/Flush (7 d)	July 2007 – July 2009
IA3A		Storage[1]	Manure basin	September 2007 – August 2009
IN4A			Lagoon	September 2007 – August 2008
NC3A			Lagoon	November 2007 – November 2009
NC4A			Lagoon	October 2007 – July 2009
OK3A			Lagoon	September 2007 – July 2009
OK4A			Lagoon	July 2007 – June 2009
CA5B	Dairy	Housing	Free-stall barn	September 2007 – February 2010
IN5B			Free-stall barn	September 2007 – August 2009
NY5B			Free-stall barn	October 2007 – November 2009
WA5B			Free-stall barn	September 2007 – October 2009
WI5B			Free-stall barn	September 2007 – August 2009
IN5A		Storage[1]	Lagoon	December 2007 – September 2009
TX5A[2]			Feedlot (housing)	December 2007 – September 2009
WA5A			Lagoon	February 2008 – June 2009
WI5A			Lagoon	July 2007 – March 2009

[1] Lagoons and manure storage basin were monitored each season for approximately 2 weeks at a time; measurement equipment was then rotated to other monitoring locations

[2] This site was a feedlot-style dairy facility where cows were housed, but since it was an open-source (like a lagoon) it fell under the storage category of the measurement campaign

Emissions measurements were taken at a total of 15 livestock barns (5 swine, 4 dairy cattle, 4 layer and 2 broiler barns and 10 manure storage facilities (5 swine lagoons, 1 swine basin, 3 dairy cow manure lagoons, 1 dairy drylot) for anywhere from 1.5 to 2.5

years, beginning in late 2007 and continuing through early 2010. Swine farm data was collected in Iowa, Indiana, North Carolina and Oklahoma for flush-type and deep-pit barns. Dairy barn data was collected in California, Indiana, New York, Washington and Wisconsin; all were free-stall dairies. For the layer chickens, measurements were taken at farms in California, Indiana, and North Carolina; all but one farm (in Indiana) used a high-rise manure management system. The remaining farm used a manure belt to remove waste from the barns. Broiler measurements were conducted by NAEMS researchers in California and by Tyson Foods in Kentucky; both farms used a litter-based barn for production.

The study measurements were coordinated by Dr. Albert Heber of Purdue University to ensure all measurements met the same quality assurance metrics and used the same measurement techniques. The time resolution of online data for NAEMS varies; for animal barns, only daily average emission factors were released whereas for manure lagoons data was released at a time resolution of 30 minutes. Additionally, the detailed co-located meteorological observations and reporting of manure characteristics was exceedingly helpful as it allowed us to better constrain the FEM, and we found that this information was not often reported in prior ammonia emissions measurements as reported in the literature, or that previous studies were not conducted over the same length of time, and so full characterization of seasonal trends in ammonia emissions was limited.

In addition to ammonia emissions, the emissions of particulate matter, hydrogen sulfide, and non-methane volatile organic compounds were monitored. Meteorological data including temperature, wind speed and direction, and pressure were also reported daily. For the lagoon storage included in the study, files with emissions observations at a time resolution of 30 minutes were released; however, in many cases, there was limited time coverage. Depending on the farm, "daily" lagoon emissions could be characterized based on data collected over the course of several hours or as little as two 30-minute periods over the course of the day. As we will discuss in the results, the limitations of data collection, particularly for lagoon storage could be problematic since overnight emissions might be very different than those observed during the warmer late afternoon.

At various points during the study, samples of feed and samples of manure from both the housing and lagoon facilities (dairy, swine, layer and broiler) under observation were collected and tested for pH (manure) and nitrogen content (feed and manure). This data was collected a few times each year. These results showed that, in general, feed nitrogen remained relatively constant throughout the year.

3.3.3 Description of Model Evaluation Process

In this work, we began by using the previously tuned values for model parameters related to mass transfer resistance (McQuilling and Adams, 2015; Pinder et al., 2004a). After comparing emission factors from the existing literature and NAEMS, it was apparent that the data from NAEMS was collected under a wider variety of conditions and showed a greater spread in ammonia emissions factors, as is shown in Figure 3.2; after observing these differences, the mass transfer resistance parameters were re-tuned, as described in Section 3.4.1.

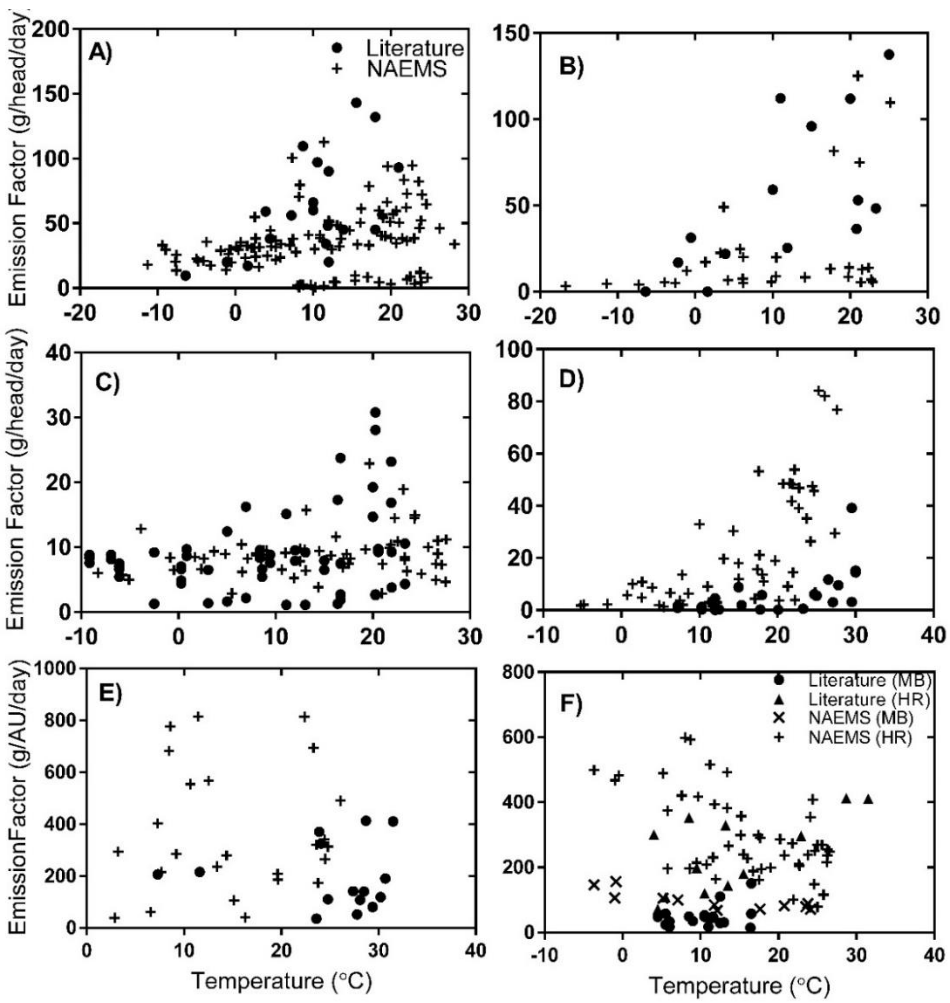


Figure 3.2: Emission factors as a function of temperature reported in the prior literature and from the National Air Emissions Monitoring Study (NAEMS). Results are displayed by animal type and management stage as follows: a) free-stall dairy housing emissions, b) dairy lagoon storage emissions, c) deep-pit and flush-type swine housing emissions, d) swine lagoon and basin storage emissions, e) litter-based broiler housing emissions, and f) manure-belt (MB) and high-rise (HR) layer housing emissions. (1 AU = animal unit = 500 kg live animal weight)

We used a variety of metrics to evaluate the performance of the model in its ability to estimate ammonia emissions. Model performance was evaluated in its correlation between model and measurement, mean fractional error, and mean fractional bias. The equations used to calculate these metrics are shown below in equations 3.1-3.3.

$$\boldsymbol{Correlation} = \boldsymbol{r} = \frac{\boldsymbol{n}\sum EF_{meas} * EF_{mod} - (\sum EF_{meas})(\sum EF_{mod})}{\sqrt{n(EF_{meas})^2 - (EF_{meas})^2} * \sqrt{\sqrt{n(EF_{mod})^2 - (EF_{mod})^2}}} \tag{3.1}$$

$$Mean\ Fractional\ Error = MFE = \frac{1}{N} * \sum_{i=1}^{N} \frac{|EF_{mod} - EF_{meas}|}{\frac{1}{2} * (EF_{meas} + EF_{model})} * 100 \tag{3.2}$$

$$Mean\ Fractional\ Bias = MFB = \frac{1}{N} * \sum_{i=1}^{N} \frac{EF_{mod} - EF_{meas}}{\frac{1}{2} * (EF_{meas} + EF_{model})} * 100 \tag{3.3}$$

The results of the model evaluation in terms of model-measurement correlation, mean fractional error, and mean fractional bias are presented in Tables 3.3. For both swine and dairy model performance evaluations, correlations were calculated and presented for daily and seasonal model performance.

3.4 Results

Previously published literature studies suggest that there is a strong relationship between ammonia emissions and the meteorological conditions, management practices, and feed nitrogen content under which those emissions occurred. Here, we have further investigated these relationships using the newly available data from the National Air Emissions Monitoring Study (NAEMS). The farm emission model that was developed based on literature (described and evaluated previously (McQuilling and Adams, 2015; Pinder et al., 2004a)) is further tested against this data. There are three major components to the NAEMS analysis in this chapter. First, we re-tune the model to better fit the data collected during the NAEMS campaign to fit the full range of conditions for which emission factors were reported by NAEMS. Secondly, the model performance is evaluated in its ability to reproduce seasonal ammonia emissions variability for dairy housing and lagoons, swine housing and lagoons, and layer and broiler housing. Since NAEMS monitored farms for multiple years and provided daily measured emission factors for the farms, we have also investigated our model's skill in reproducing day-to-day variability in ammonia emissions. This was a stringent test of the model's skill since it was originally developed to capture seasonal differences in emissions, not necessarily day-to-day variability.

3.4.1 Synthesis of existing literature and NAEMS data

The FEM was originally tuned solely to data gathered from literature predating the NAEMS study (McQuilling and Adams, 2015). With the advent of the NAEMS data, one approach would be to test the previously tuned model against the novel and wholly independent NAEMS data. However, as will become clear below, the NAEMS data goes substantially above and beyond what was previously available for model tuning, exploring emissions under conditions that were not sampled previously, and in some cases, looking qualitatively different. It was apparent that predicting NAEMS measurements under conditions that were not sampled previously is essentially an extrapolation of the model and unlikely to be informative or successful. Therefore, it was decided to re-tune the FEMs to the full suite of available data, including both NAEMS and the earlier literature measurements. The model evaluation that follows, therefore, does not reflect the ability of the FEMs to predict completely independent measurements but the ability of a relatively simple process-based model, with a single set of mass transfer parameters for each manure management practice, to describe the full range of observed variability.

The NAEMS data and literature data are displayed previously in Figure 3.2. The range of temperatures studied is most extended for layer hens. With the additional NAEMS data, an apparent inverse relationship between temperature and ammonia emissions is observed, something that was not clear in the prior literature. It has been suggested that this inverse relationship (higher emissions factors for lower temperatures) is related to the drying out of manure in hot barns with high ventilation rates (Morgan et al., 2014). At lower temperatures, barn ventilation is reduced (to conserve heat) and manure dries slowly, and, therefore more manure urea can be broken down into ammonia, which is then available for volatilization. Additionally, we saw that for some practices, particularly for swine storage, emissions factors from NAEMS were uniformly higher than those previously reported in the literature, for both high and low temperatures. As a result of these differences, the FEM's tuned parameters were adjusted so that model emission factors fell between NAEMS and literature data, weighting the literature studies equally with the NAEMS observations so as not to over-tune to only the literature or NAEMS data. There is significant value in both previously published studies as well as in the values reported by NAEMS, so the model recalibration done is to ensure that this work takes advantage of all available data. As we were trying to capture the emission factor variability observed both in the literature and in the NAEMS observations, the process of recalibration was less formal (than the original tuning of parameters), and the model was adjusted to produce results in between the results from the literature and NAEMS if they were substantially different. When the NAEMS observations and literature did not differ by a large amount, the changes between pre- and post-recalibration were minimal, while for some farm types (namely swine storage and layer housing) the re-calibration produced results that were markedly different. Figure S3.1 in the supplemental figures shows literature emission factors, NAEMS emission factors, and emission factors from the model run for the NAEMS farms pre- and post- recalibration for the animal types which had the greatest differences between the literature emission factors and NAEMS observations, swine storage and layer housing.

3.4.2 NAEMS and performance in capturing seasonal variability

Figure 3.3 presents, for six NAEMS farms, the measured seasonal cycle of emissions compared to that predicted by the corresponding FEM. The results in Figure 3.3 are typical (the farm that is, on average, closest to average mean fractional error of the farms in the study) of each animal type and manure management stage included in NAEMS: dairy housing, dairy (lagoon) storage, swine housing, swine (lagoon and basin) storage, broiler housing and (high-rise and manure-belt) layer housing.

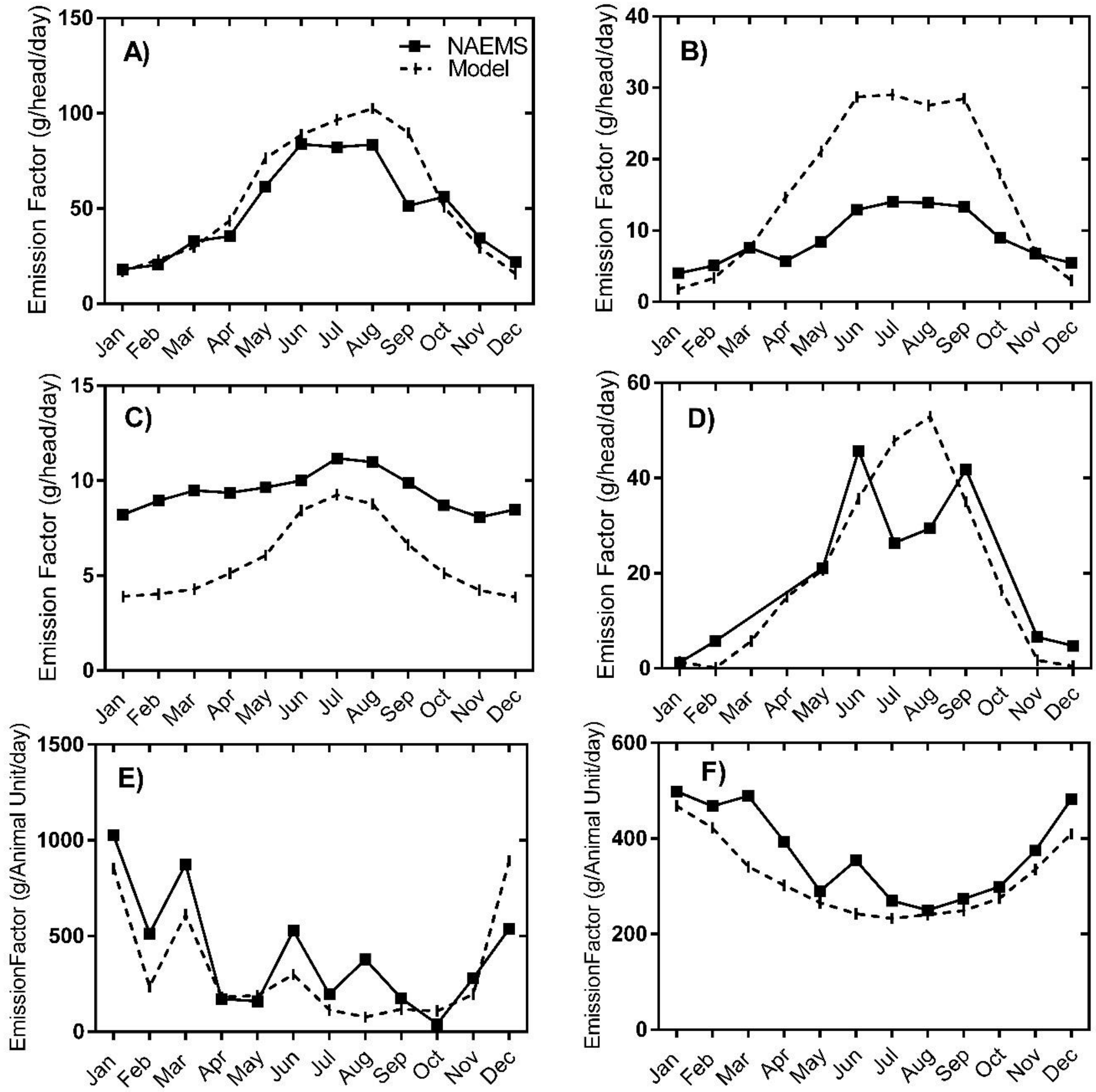


Figure 3.3: Comparing seasonal model performance for each animal type and management stage. Animal types and stages are as follows: a) free-stall dairy housing in NY (NY5B), b) dairy storage lagoon in IN (IN5A), c) shallow pit swine housing in NC (NC4B), d) swine storage lagoon in OK (OK3A), e) litter-based broiler housing in CA (CA1B), and f) high-rise layer housing in IN (IN2H). Farm evaluations for all sites in the study are included in the Supplemental information in Figures S3.1-S3.6.

Specific details about each of the farms studied are presented in Table 3.2 including when the study occurred. The remaining farms are shown in the supplemental information in Figures S3.2-S3.7.

Table 3.3: Errors and correlation coefficients for daily and seasonal model performance

Animal Type	Farm	Management Stage	Seasonal Correlation	Daily Correlation	Seasonal Mean Fractional Error	Seasonal Mean Fractional Bias	Daily Mean Fractional Bias
Dairy	CA5B	Housing	0.62	N/A	170%	85%	N/A
	IN5B	Housing	0.95	0.59	18%	0%	-35%
	NY5B	Housing	0.87	0.3	19%	2%	1%
	WA5B	Housing	0.03	0.25	47%	-1%	-84%
	WI5B	Housing	0.96	N/A	13%	6%	N/A
	IN5A	Storage	0.94	0.31	59%	29%	12%
	WA5A	Storage	0.59	-0.76	39%	94%	55%
	WI5A	Storage	0.88	-0.04	90%	-90%	-42%
Swine	IA4B	Housing	0.78	-0.01	21%	-13%	-102%
	IN3B	Housing	0.34	0.49	44%	-4%	24%
	NC3B	Housing	0.09	0.01	21%	7%	22%
	NC4B	Housing	0.35	0.42	23%	18%	42%
	OK4B	Housing	0.91	0.25	17%	-10%	-3%
	IN4A	Storage	0.77	0.04	22%	-14%	8%
	NC3A	Storage	0.83	0.23	15%	-7%	103%
	NC4A	Storage	0.94	0.42	84%	42%	-47%
	OK3A	Storage	0.82	0	91%	-41%	9%
	OK4A	Storage	0.68	0.09	35%	-3%	-89%
Layers	CA2B	Housing	0.87	N/A	27%	-4%	N/A
	IN2B	Housing	0.84	N/A	16%	13%	N/A
	IN2H	Housing	0.89	N/A	16%	-8%	N/A
	NC2B	Housing	-0.43	N/A	28%	25%	N/A
Broilers	CA1B	Housing	0.5	N/A	60%	-20%	N/A
	KY1B	Housing	0.61	N/A	61%	-5%	N/A

Note: Not all months of study were evaluated for each farm for daily model performance; a warm-weather period and cold-weather period was evaluated for each farm. Results may or may not be entirely representative of daily model performance over the longer term.

As shown in Figure 3.3a-3.3d, dairy cattle and swine farms typically have summer peaks in emission factors, while layers more commonly have winter peaks, as described in Section 3.4.1 (Morgan et al., 2014). The emissions factors for broiler farms tend to be driven more by animal size, because of their brief life cycle (only 6-8 weeks on most farms), though there is also a seasonal cycle. In our model, because of the data reported by NAEMS about animal age, we are able to adjust for animal age and weight because older and larger animals are able to more efficiently use the nitrogen they are fed. Here, for purposes of comparing with a specific farm where animal age is known, we included the average age of the animals in each month as an input as a way to adjust for differences in nitrogen use efficiency. We anticipate that, for purposes of building emissions inventories, when one is summing across numerous barns or farms in a county, these farms will have their animals in different stages of the life cycle; therefore, these adjustments will tend to average out in emissions inventories and can be ignored, leaving only a very weak seasonal trend in emissions at the inventory level.

The results of the seasonal model evaluation for dairy housing and storage are shown in Table 3.3. In general, the model performs reasonably well in terms of correlation coefficient for seasonal variability, ranging from 0.62-0.96 (with the exception of the Washington barn where $r = 0.03$, whose seasonal model performance is shown in Figure S3.2d and whose measured seasonal cycle was atypical for unknown reasons). In terms of mean fractional error and bias (excluding the California barn, which has observed emission factors approximately one-tenth of those seen in other study locations measured under similar conditions), MFE ranged from 18%-47% while MFB was between -1% to 6%. Thus, if we modified the model to better capture the seasonal variability for the farm in Washington, the model would no longer adequately describe the seasonal trends in emissions for the other dairy farms included in NAEMS. This discrepancy highlights the role of farm-to-farm variability in ammonia emissions factors and the fact that there are other factors contributing to this variability not captured by the FEM described previously (McQuilling and Adams, 2015; Pinder et al., 2004a).

The results of the seasonal model evaluation for swine housing and storage are shown in Table 3.3. In general, the seasonal correlations are better for the lagoons (manure storage) than for barn emissions, indicating that the lagoon emissions are most strongly driven by temperatures and vary most greatly across seasons. The seasonal correlations range from 0.09 to 0.91 for housing and 0.68 to 0.94 for swine manure storage; the lowest correlation is for farm NC3B, which shows lower than expected summertime ammonia emissions; other farms have emissions factors up to an order of magnitude higher for similar meteorological conditions. In contrast, the mean fractional error and biases in the measurements are substantially greater for the open sources measured during NAEMS. This is likely related to the additional measurements, analyses, and assumptions required to infer emissions from outdoor sources compared to indoor housing with well-known ventilation rates. These additional steps include the monitoring of upwind and downwind concentrations, frequent measurement of wind speed in order to calculate emissions trajectories, and the use of a dispersion model to calculate emissions based on these factors. Mean fractional errors for animal housing were modest, ranging from 17%-44%, whereas they could be much higher for outdoor storage: 15%-91%. Similarly, mean fractional biases for swine housing ranged from -13%-18% and storage ranged from -41% to +42%.

Model evaluation for seasonal model performance for poultry is also shown in Table 3.3, including both layer farms (CA2B, IN2B, IN2H, and NC2B) as well as broiler farms (CA1B and KY1B). For the layers, two types of housing were considered: high-rise (CA2B, IN2H, and NC2B) and manure-belt (IN2B). The main difference in these housing options is the frequency at which the manure is removed: every several months for high-rise houses and multiple times per week for manure-belt houses. For the layers, the model performed reasonably well on a seasonal basis for 3 of the 4 farms. The NC2B farm, a high-rise barn, showed the opposite seasonal trend in emissions when compared to the other three farms. The correlation for the other three layer farms ranged from 0.84-0.89, whereas for NC2B it was -0.43. Across all layer housing, mean fractional errors ranged from 16%-28% and mean fractional biases between -8% to 25%. Only two broiler farms were included in NAEMS. The model evaluation for these two farms is also shown in Table 3, where r ranges from 0.5 to 0.61 and mean fractional error is ~60%. Mean fractional bias was between -5% to -20%, which reflects that many previous studies observed lower ammonia emissions factors than those seen in NAEMS.

3.4.3 Evaluation of daily variability

One of the unique features of the National Air Emissions Monitoring Study was its large set of daily emissions factors for ammonia. Because this data was available, and because of the role of ammonia emissions in episodic formation of particulate ammonium nitrate, especially during the wintertime (Stanier et al., 2012), we wanted to see if our model could capture the daily variability seen in the ammonia emissions observations from NAEMS. Results of this analysis are shown in Figures 3.4-3.5 and Table 3.3 and the supplemental information in Figures S3.8-S3.11.

Overall, the results from the daily evaluation of the dairy and swine FEMs (originally developed to capture the seasonal variability in ammonia emissions) are mixed but encouraging. As seen in Figure 3.4, as well as in Table 3.3, a modest amount of daily variability is captured at some farms but not for others—for 8 of the farms the correlation coefficient is greater than 0.25, for 7 additional farms the correlation coefficient is between -0.04 and 0.23, and for one dairy lagoon, the correlation coefficient is -0.76.

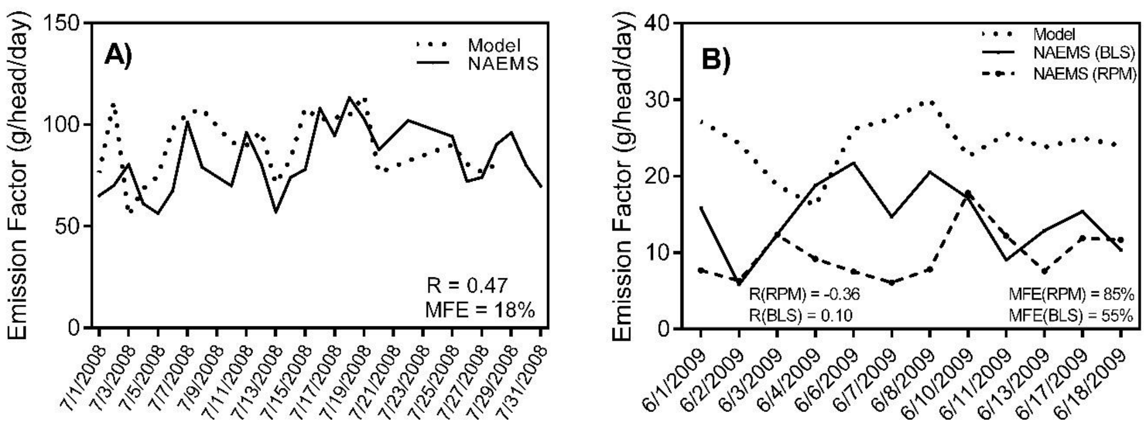


Figure 3.4: Daily variability in NAEMS data and model predictions for: a) a free-stall dairy house in Indiana during July 2008 (IN5B) and b) a dairy lagoon in Indiana during June 2009 (for both RPM and BLS methods of emission factor estimation) (IN5A). Additional daily evaluations are found in the Supplemental information in Figures S7-S10.

Typically, the daily model-measurement correlation is better for housing, but not always; in general, the worst housing correlation is better than the worst storage correlation. Also, as evidenced in Figure 3.4b, the two methods used by NAEMS to infer lagoon emissions (RPM = ratiometric plume mapping and BLS = backwards lagrangian stochastic modeling) produce significantly different results. The daily correlation coefficient between the RPM and BLS measurements can range dramatically from farm to farm, varying from -0.8 to nearly 1. This suggests that some of the apparent failure of the model to reproduce daily variations stems from measurement errors themselves. Future inventory improvements will require the development of more robust measurement methods to constrain the models and inventories.

Model performance can also be judged qualitatively, whether it captures a significant portion of the day to day observed variability in emissions (R>0.25), whether it performs neutrally (neither good nor bad, -0.04<R<0.25), or whether it performs poorly (R<-0.04). By these standards, the model performs well for 44% of the swine and dairy houses and lagoons, neutrally for 44% of the swine and dairy farms, and performs poorly for the remaining 12% of farms.

Day-to-day variability in poultry emissions was not well captured by the FEM. In Figure 3.5, the differences between seasonal trends and day-to-day trends with respect to temperature are shown for the farm CA2B, a high-rise layer barn in California.

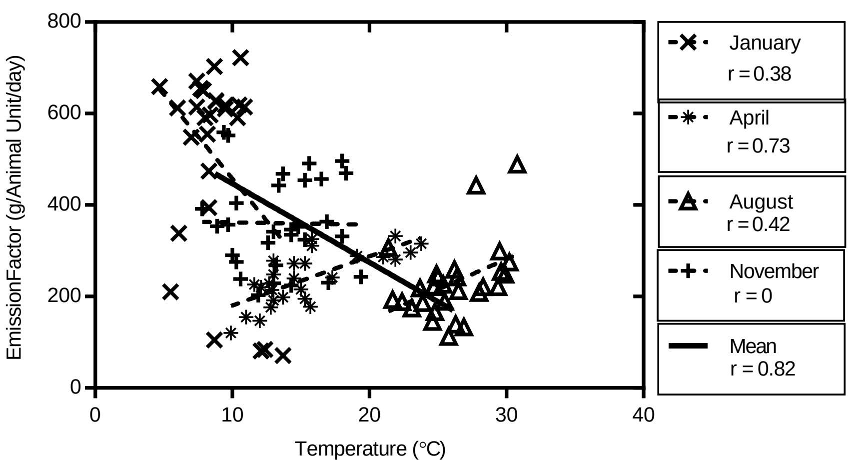


Figure 3.5: Comparison of daily and seasonal variability in emission factors as a function of temperature for a high-rise layer chicken house in California monitored during NAEMS

Here, on a seasonal average, higher emissions occur in winter and thus associated with lower temperatures; similar results are observed for the layer farms in Indiana. In contrast, for daily variability in emissions, within a given month, often, though not always, a positive emission correlation with temperature, or even no relationship between temperature and ammonia emissions at all. Given the current FEM structure and the lack of physical explanation for these trends, it was not possible to construct an FEM that simultaneously captured seasonal and daily variability. As a major goal of the FEM is to use it to construct a new national inventory and given the importance of the seasonal variability for predicting fine PM formation, we opted to maintain the model's tuned parameters given that they perform well for seasonal variability. This indicates that our simple semi-empirical process-based emission model does not include all the factors and processes that may affect ammonia emissions at the day-to-day level of time resolution.

3.4.4 Farm-to-farm variability

In addition to capturing seasonal and daily variability in emissions, it is important to ensure that the FEMs reproduce farm-to-farm variability that results from regional differences in climatology as well as farm-to-farm variations in manure management practices and animal feeding. For each of the farms modeled, we calculated an annual average emission factor based on both the observations and model results, seen in Figure 3.6.

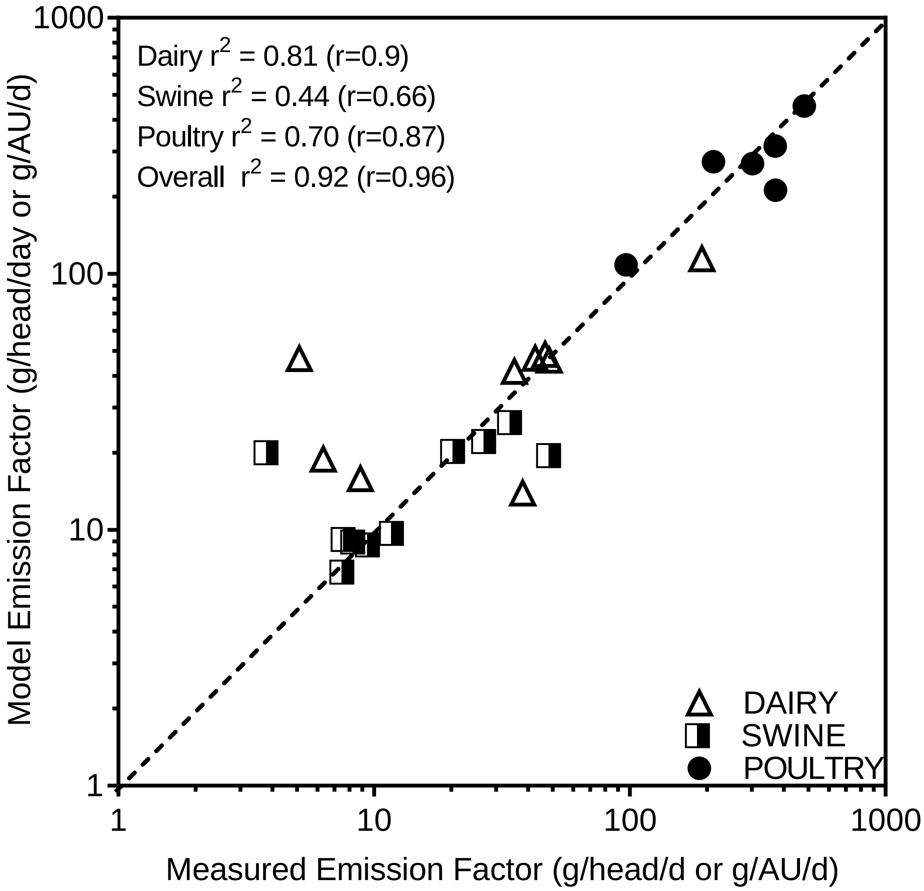


Figure 3.6: Comparison of modeled versus measured average annual emission factors for all farms monitored during NAEMS (dairy, swine, and poultry farms included).

Figure 3.6 shows the model's annual average emissions factors compared the measured annual average emissions factors, with an overall r^2 value of 0.92 (R value is 0.95). If the data are separated by animal type (dairy, swine and poultry), the r^2 values are 0.81, 0.44, and 0.70 for dairy, swine, and poultry respectively (R values are 0.9, 0.66, and 0.84 for each animal type). If we further separate the farms by not only animal type but by grouping also by the specific practice (and associated mass transfer resistance parameters), results are slightly more mixed. This information is presented in Table 3.4.

Table 3.4: Annual Average Emissions Factor model performance by animal and farm type

Animal Type	Farm Stage	Type	Number of farms of this type studied in NAEMS	Valid Comparison Possible?	Farm to farm T variability as fraction of average T range	% EF Variability (relative to Average EF)	Model vs Measurement R value
Swine	Housing	Deep pit	2	No	n/a	n/a	n/a
		Shallow	3	Yes	0.14	0.05	-0.99
	Storage	Lagoon	5	Yes	0.23	0.31	0.18
	Storage	Basin	1	No	n/a	n/a	n/a
Dairy	Housing	Freestall	5	Yes	0.35	0.15	0.06 (0.84 excluding CA5B)
	Storage	Feedlot	1	No	n/a	n/a	n/a
	Storage	Lagoon	3	Yes	0.14	0.31	-0.83
Layer	Housing	High-rise	3	Yes	0.25	0.16	0.78
	Housing	Manure Belt	1	No	n/a	n/a	n/a
Broiler	Housing	Litter-based	2	No	n/a	n/a	n/a

Some of the comparisons are difficult or impossible to make owing to how few farms fall into certain categories (e.g. layer manure-belt housing, swine deep-pit housing, swine basin storage, broiler housing, or a dairy cattle feedlot housing/storage). Additionally, it was difficult to evaluate model performance where there was little meteorological variability between farm locations, for example, all the shallow-pit swine farms had similar meteorology as well as little difference in annual average emission factors. In this case, the farm-to-farm differences are not able to be separated from other sources of variability in emissions factors not captured by the FEM. The model performed best when there were at least 3 farms of a particular type that were located with significantly different average meteorology and more widely varying emission factors. With these greater ranges in meteorology and emissions, the model is better able to characterize farm to farm differences. The performance evaluation shown in Table 3.4 indicates that some of the overall skill of the FEM is simply due to its separation of different farm practices by using different mass transfer resistance parameters. Overall, the strengths of the FEMs are largely in their abilities to do capture seasonal and daily variability in emissions and investigate “what-if scenarios” in which different management choices are considered.

The mean fractional error for each animal type, with the exception of the farms whose emissions were drastically different in magnitude than the other farms for similar conditions, are: 31% for dairy farms, 18% for swine farms, 17% for layer farms, and 25% for broiler farms. These fractional errors typically fall within the range of uncertainty in measurement. As discussed in Chapter 2, the measurement of ammonia emissions can be difficult, especially for outdoor (open) sources; for instance, emissions estimates from the use of backwards Lagrangian stochastic models is estimated to be 15-20% or greater (up to more than 100%) for unstable conditions (Flesch et al., 2004). Furthermore, there is uncertainty in our estimates due to the simplicity of the model and the fact that not all sources of variability are captured by the farm emission model.

Broadly speaking, the results in Figure 3.6 show that model results are consistent with the average NAEMS observations. Earlier inventories often treated emissions with a constant emissions factor given a particular set of practices; though this may capture some of the emissions variability resulting from different management decisions, it fails to capture the seasonal distribution of emissions, which is especially important for understanding the air quality implications of these emissions. The most recent National Emissions Inventory (2011 NEIv2) has added a temperature dependence to the ammonia emissions thus capturing some of the variability not previously addressed (USEPA-OAQPS, 2015a).

3.5 Conclusions

The National Air Emissions Monitoring Study (NAEMS) offered a new source of data for us to use to improve and evaluate our Farm Emissions Models (FEMs). In order to model the ammonia emissions from the NAEMS farms and capture their observed variability (as a result of meteorological differences and differences in practices), we need to use a semi-empirical process-based model which can account for these differences, often left unconsidered in previous inventories. We evaluated the FEM against data from NAEMS for dairy housing and lagoons, swine housing and lagoons, and layer and broiler housing in terms of the model's ability to capture seasonal, daily, and farm-to-farm variability. Data used from NAEMS include meteorological parameters, feed nitrogen content, and manure pH.

The model performed well in capturing seasonal variability in emissions factors—for all but one of the dairy barns, the correlation coefficient ranges from 0.62 to 0.96; the remaining farm, a freestall barn in Washington, has an R-value of 0.03 and an unusual measured seasonal cycle. Swine housing results had R-values ranging from 0.09 to 0.91, with an average value of 0.5. Seasonal correlations for both dairy and swine storage ammonia emissions were quite strong with R-values of 0.59 to 0.94 and an average of 0.81. Mean fractional errors for swine and dairy housing were typically 10%-47% with biases mostly less than +/-20%. Storage emissions showed a greater range in error and biases, which could be due to model error or difficulties associated with conducting outdoor ammonia emissions measurements, which require a dispersion model to calculate emissions based on observed concentrations. Similarly, for layer housing, three of the four farms have a model-observation seasonal correlation of between 0.84-0.89, but the farm with different seasonal characteristics, a high-rise layer farm in North Carolina, and the r value is -0.43. This result highlights the farm-to-farm ammonia emissions variability that can be observed, even on farms using the same practices and using the same techniques for emissions measurements. The seasonal correlation coefficients for broilers were 0.50 and 0.61 respectively.

The FEMs ability to predict daily variability in emissions shows promise but was less strong overall than the seasonal evaluation. For half of the farms evaluated, the daily correlation coefficient is greater than 0.25, demonstrating that a component of the daily variability can be explained simply with daily variation in meteorology. For most of the remaining farms, the model had little or no skill in predicting daily variability (daily correlation coefficients between -0.04 and 0.23), and for one dairy lagoon, the correlation coefficient is -0.76. Generally, the daily model performs better for housing than storage. The apparently poor FEM performance in predicting daily emissions from storage may result partly from the difficulty of performing these measurements, accounting for dispersion, and inferring emissions. For example, even two widely used methods for computing emissions, RPM and BLS, do not always correlate well with each other on the daily time scale. Additionally, we found that daily variations in ammonia emissions for poultry were not well captured by the FEM; for three of the four layer farms, seasonally there were higher emissions associated with colder temperatures while in terms of daily variability, warmer temperatures tended to be associated with higher emissions.

In addition to evaluating model performance in terms of seasonal and daily ammonia emissions factors, we also wanted to be sure the FEM was able to differentiate between farms and practices. To do this we compared the average annual emission factor for each farm monitored from the FEM and the NAEMS data. As shown in Figure 3.6, the overall r^2 value is 0.92 for all animal types, while, when considering dairy, swine and poultry separately the r^2 values are 0.81, 0.44 and 0.70 respectively. This result shows the model's skill in capturing big picture emissions as well as the ammonia emissions variability driven by practices in addition to meteorology which has been shown in both seasonal and daily evaluations. Our model is relatively simple, as it relies on accessible input data (meteorological conditions) and a few tuned parameters and so cannot be expected to capture all the variability in emissions; this is especially true for open-sources (e.g. lagoons) whose emissions can be highly uncertain, even using most recent measurement techniques.

This chapter has demonstrated the FEMs' skill in capturing the seasonal, daily and farm-to-farm variability in ammonia emissions as observed during the NAEMS campaign. The data collected during NAEMS provided an unprecedented level of detail in terms of farm conditions and long-term observations of emissions that allowed us to improve the model's performance. Chapter 4 details how the FEM is used as the basis for a new national inventory for cattle, swine and poultry ammonia emissions.

CHAPTER 4: DEVELOPING A NEW NATIONAL INVENTORY--DATA SOURCES, MANAGEMENT PRACTICES AND NH_3 EMISSIONS

4.1 Abstract

The prediction of $PM_{2.5}$ concentrations in chemical transport models (CTMs) requires emissions inventories with spatial and seasonal variation. One promising approach for the creation of this type of inventory is a process-based model. Here we apply the results presented in the previous chapters to the US populations of swine, cattle, and poultry. Temperature, wind speed and precipitation data were used for 2011; future work will construct an updated inventory for 2014 with its own meteorological inputs from the National Climate Data Center. Regionally-specific manure management practices were obtained from the most recently completed National Animal Health Monitoring Surveys (NAHMS) for beef, dairy, swine, and poultry; the survey data used is all from 2006-2012. Animal populations by county were collected from the most recent USDA animal census, completed for 2012.

Based on our model, total annual ammonia emissions for 80 million beef cattle, 9 million dairy cows, 65 million swine, 350 million layers and 1.5 billion broilers are estimated to be 720 Gg, 202 Gg, 435 Gg, 93 Gg, and 264 Gg respectively, totalling 1.7 Tg of ammonia emissions annually; detailed results from this work are described in this chapter. The most recent National Emission Inventory (NEI2011v2) had livestock ammonia emissions totalling 1.97 Tg, which is greater than our inventory, but the limited characterization in the literature of some manure management practices introduces additional uncertainties to our model's results. Comparing to the inventory previously

produced by Pinder et al. (2006), our results are approximately 220 Gg lower annually, a difference of less than 20%, which is within the range of uncertainty in our model.

Some regions' ammonia emissions are driven by a single livestock type, while other locations are heavily agricultural and have significant emissions from cattle, swine, and poultry. As expected, ammonia emissions vary seasonally, typically with a greater emissions in the summer than the winter, but locations dominated by poultry tended to have a weaker dependence on meteorology than those dominated by cattle or swine emissions. Additionally, significant day to day emissions variability is noted estimated by the model; total ammonia emissions in the United States can vary by more than 1500 tons from one day to the next. Warmer days during the winter appear to have a particularly strong effect on the emissions factors produced by the model than those during the summer, which could particularly important for particulate matter formation, especially in the Midwest (Stanier et al., 2012).

4.2 Introduction/background

4.2.1 Previous inventories

Emissions of ammonia are highly seasonally variable, as discussed in previous chapters. Additionally, the role of ammonia in particulate matter formation is also seasonally variable; during the wintertime, excess ammonia is more likely to react with nitric acid to form particulate ammonium nitrate, as this reaction is favored at colder temperatures. This reaction is less thermodynamically favorable during the summer with warmer temperatures. As a result of the differences in the role of ammonia in particulate matter formation during different seasons, it is particularly important that the emissions inventory used captures these seasonal differences in order to be used to accurately predict fine particulate matter concentrations throughout the year via the use of a chemical transport model (CTM).

Previously, there have been a number of approaches to estimate ammonia emissions at various spatial and temporal resolutions in order to produce a national level ammonia emissions inventory. Traditional inventories (Battye et al., 2003) have most often relied on the use of single emission factors for each animal type to characterize emissions; this approach captures none of the variability in emissions that results from differences in manure management practices and meteorology that are known to impact ammonia emissions. More recent national inventories (USEPA-OAQPS, 2015a), like the NEI 2011v2 have implemented a temperature dependence, where emissions in a particular location are a function of the daily average temperature (with higher emissions in the summer and lower emissions during the winter), in to the ammonia emissions inventory for livestock. While this method is able to capture a significant portion of emissions variability from differences in temperature, it still fails to account for the emissions differences resulting from regionally-specific manure management practices and the other environmental factors (besides temperature) that can impact emissions of ammonia from housing, storage, and manure application from livestock production. Several emissions inventories for ammonia emissions in Europe have employed a more process-based approach, but practices in the United States often differ substantially from those employed throughout Europe (Hutchings and Sommer, 2001; Li et al., 2012; Misselbrook and Chadwick, 2010; Webb et al., 2005).

4.2.2 Goals of the process-based inventory

The inventory described in this chapter accounts for both the emissions variability resulting from manure management choices as well as meteorology (temperature, wind speed, and precipitation) in a manner similar to that developed for dairy cows previously (Pinder et al., 2004b), utilizing the farm emissions model (FEM) framework described in Chapters 2 and 3. This earlier inventory for dairy cows was developed to capture the seasonal variability in emissions. Here, we wanted to develop inventories not only for dairy cattle, but also beef cattle, swine, and poultry (layers and broilers), by including all major livestock types we create a more complete picture of national ammonia emissions. Additionally, this inventory is different than previous work because it produces daily emissions of ammonia; this level of temporal resolution was evaluated using data from the highly time-resolved data (from NAEMS) and described in Chapter 3. The goal of this work was to produce an inventory of ammonia emissions with daily time resolution for beef, swine, and poultry (layers and broilers) in the United States.

This chapter describes the construction of the ammonia emissions inventory. Information about animal population, distributions of manure management practices, daily time-resolved emission factors, total ammonia emissions by county and by animal type based on an assumed distribution of practices, and a discussion of the sensitivity of the model and inventories to the regional distribution of manure managment practices for beef, dairy, swine, and poutlry in the United States are all included.

4.3 Data sources and Method Description

4.3.1 Meteorological data

In order to produce a daily time-resolved emissions, the FEMs require daily average temperature and wind speed as well as daily precipitation as inputs. The inventory produced previously (described in Pinder et al. (2006)), took as inputs from the National Climate Data Center (NCDC) the long-term monthly average temperature, monthly average windspeed and total monthly precipitation. Then, the FEM computed a statistical representation of day-to-day temperatures using the standard deviation in temperature within a given month. There is no wind variability in day to day wind speed; the monthly average wind speed is used for every day in the month. For precipitation, the precipitation frequency to be used in the model was defined by a single input. This allows for days with and without precipitation but does not mimic actual precipitation patterns because each precipitation event was of the same magnitude and occurred with the same frequency.

In this inventory, produced for 2011, we retrieved actual daily average temperatures, windspeeds, and daily precipitation totals (rather than climatological estimates of typical meteorological conditions) from the NCDC. There are 345 climate divisions in the United States, and we used daily temperature, wind, and precipitation data for each of these. Some of the climate divisions had more than one site at which all required data was collected. In the event that there was more than one site in a climate division, the site with the most complete data was selected. If there was more than one site with complete temperature, wind and precipitation data for 2011, the site with the location most central to the climate division was selected. After selecting the sites to be used as representative of each of the climate divisions, the meteorological parameters were reviewed to make sure that all days had a reasonable value; missing data was denoted as -9999 in the documentation; if there were missing daily values for temperature or wind speed, the previous day's temperature or wind speed was used. For missing precipitation entries, we assumed precipitation on that day to be zero. For some climate divisions (~20 divisions), there were no sites within the climate division boundaries that had wind speed data available. For these locations, where there was no wind speed data available, the the wind speed from the nearest climate division was used.

4.3.2 Animal Populations

County-level animal population data is taken from the USDA Animal census. The census is taken every 5 years, most recently in 2012. We have assumed that there were no significant changes in the animal populations for any of the animal types between 2011 (the year for which the inventory is being produced) and 2012 when the census was conducted. There may be small fluctuations in livestock population, but since the annual data is only resolved at the state level, we are unable to accurately adjust county-level populations from the 2012 census, so we assumed that animal populations in 2011 were equivalent to those in 2012. Additionally, we plan to construct an ammonia emission inventory for 2014 using the same methods, and so we have compared state populations from 2012 to those from 2014. Generally speaking, animal populations did not change significantly, typically less than a 5% difference in state-level animal populations between 2012 and 2014. We have population data at the county-level for the following animals: dairy cows, beef cows, feedlot beef cows, swine, layer chickens, and broiler chickens.

Additionally, there are some counties for which data is unavailable; for these, we assumed that the missing counties had a population equal to the average "missing farm." This calculation is described in equation 4.1 below.

$$Missing\ county\ population\ = \frac{Total\ State\ population - \sum_{i=1}^{\#\ counties\ w/data} population_i}{number\ of\ missing\ counties}$$

Note: There are two sets of population numbers used in these calculations, the state total population and the county populations. The difference between the state total and the sum of the individual county populations is distributed across the counties in that state for which the animal population is unspecified.

The same approach is used for missing county-level animal populations for all states and animal types; this may cause certain counties to be over or underestimated, but should average out at the state and regional level. Additionally, for states where the total population is unknown, the missing counties are assumed to have a population of 0 for that animal type/practice. As a result of this missing population data, our inventory for these locations will obviously be biased low. This is expected to have little effect on the overall inventory; state total populations are only unknown for 1 state for swine and 4 states for beef feedlot emissions. The missing state has less than 0.1% of the total swine population in the country and the 4 states without feedlot population totals make up less than 2.4% of the total beef cattle population in the United States (of which less than 15% is expected to be housed on feedlots).

4.3.3 Regional Manure Management Practices

4.3.3.1 Overview

In addition to location-specific meteorology, our model requires farm-type inputs which describe the type of animal housing, manure storage and application methods used for a particular location. Each location is expected to have some combination of practices; for example, in a single county, some of the swine farms may use deep-pit housing, lagoon storage, and irrigation application while other farms use shallow-pit housing with lagoon storage and injection application. In order to understand the differences in regional preferences for particular manure management strategies, information was extracted from the most recent National Animal Health Monitoring Surveys done by the USDA. The beef cattle NAHMS was completed in 2007 and feedlot beef in 2011; dairy cattle data was from 2002 and 2007; swine data were collected for 2006 and 2012, and the most recent poultry NAHMS was completed for 2010. The most recent data available had limited spatial resolution (compared to previous work (Pinder et al., 2004b)), and so this work is only able to resolve large-scale regional differences in practices. For beef cow-calf systems, the United States was divided into four regions, but only two regions for beef housed on feedlots. For swine, the country was divided into three regions—Midwest, East, and South, and for layers, there were four regions—Northeast, Southeast, Central and West. An additional limitation in the data available for the characterization of the farm practices was that for some of the questions asked by the study, results were only reported in terms of percent of operations which used a particular practice. This may give too much weight to the practices used on smaller farms which have a relatively small contribution to the overall level of ammonia emissions from a particular livestock type or practice. Thus, some uncertainty is expected as a result of the limited quantity of data available regarding manure management practices throughout the country.

As was previously discussed by Pinder et al. (2004b), one of the factors most limiting to the FEM's skill is the lack of information about manure managment practices throughout the country. It is unclear whether these uncertainties result in the overprediction or underprediction of total ammonia emissions from livestock in the United States.

4.3.3.2 Dairy Practices

The distribution of practices used in dairy cattle is unlikely to have changed substantially in the years following the work of Pinder et al. (2004a, 2004b), as seen when comparing the two most recent NAHMS results (from 2002 and 2007) to the 1996 NAHMS data used in the cited work. However, the data available for the 2002 and 2007 NAHMS was less regionally specific than was used in the previous work (USDA-APHIS, 2007a, 2007b, 2007c, 2002a, 2002b). The manure management practice information received at that time included state-specific data, something not available for the current study years. Addtionally, storage and application data for 2002 and 2007 was only available by fraction of surveyed operations rather than by population which may give too much weight to practices employed primarily at smaller dairy farms. Manure management practices can be described regionally as either in the West or East; the distribution of practices is shown below in Figure 4.1a-b.

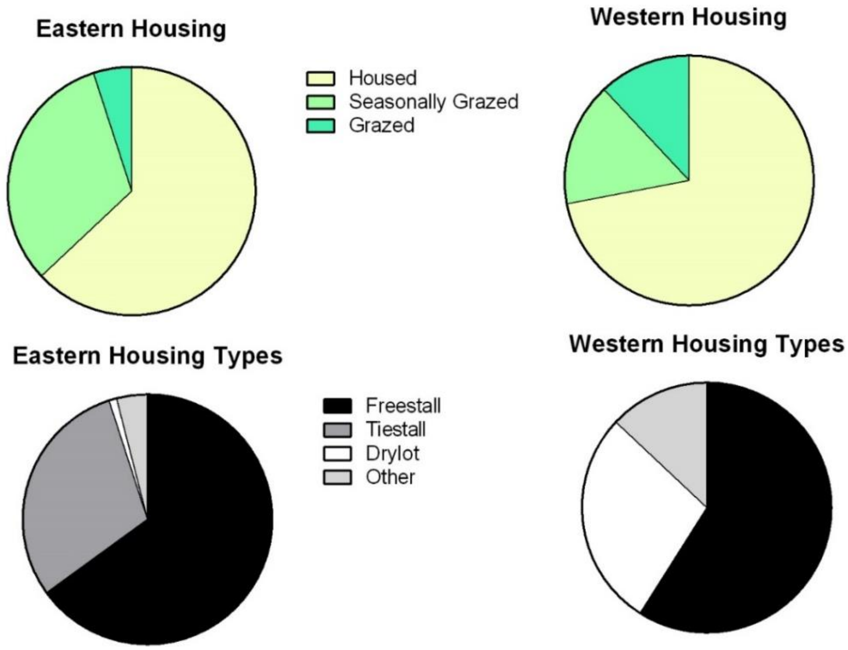


Figure 4.1a: Regional distribution of dairy housing practices from 2007 NAHMS for Eastern and Western United States. Eastern States include MN, IA, MO, AR, LA and eastward. Western states are the rest of the continental US.

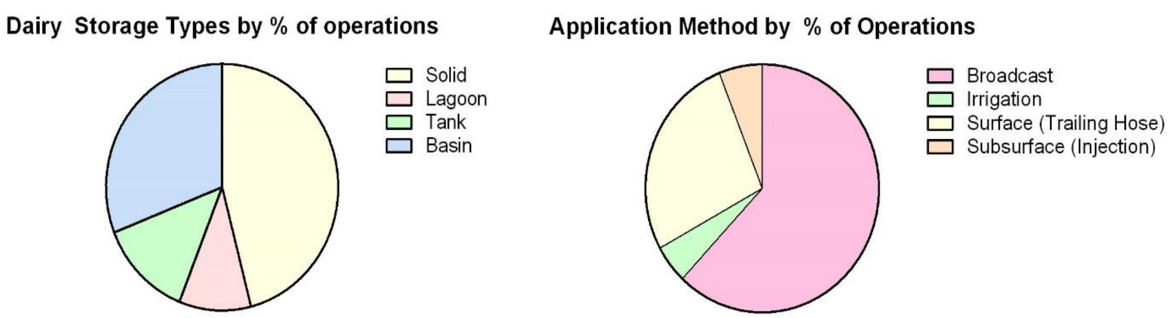


Figure 4.1b: Distribution of storage and application practices across the US. Regionally separated data was not available from the 2007 NAHMS, and results are presented in terms of % of farming operations rather than % of animal population.

4.3.3.3 Beef Practices

As stated previously, information regarding beef manure management practices was provided through the USDA National Animal Health Monitoring Study (NAHMS) with a regional distribution of practices. Beef data was provided for beef housed on feedlots as well as those that are a part of cow-calf systems. Cow-calf systems are those in which cattle are left on pasture or rangeland and the cows are kept with their calves, often until the calves are 1-2 years old and ready for sale.   Feedlots are a much denser style of production in which large numbers of cattle are housed on concrete or packed earth lots and fed a mixture of corn and grains. From the information from NAHMS and the animal numbers in the USDA 2012 agriculture census, we were able to discern the fraction of cattle in each state that were housed on feedlots as opposed those raised in a pasture-based farm system.

The distribution of manure management practices for the states included in the National Animal Health Monitoring System (NAHMS) (as split between feedlots and cow-calf systems) can be seen in Table S4.1 in the supplemental information (USDA-APHIS, 2013a, 2013b, 2009a, 2009b, 2009c). The regional distribution of cattle on feed can be seen in the Figure 2 below. There have been relatively few studies that have characterized the emissions from cow-calf or pasture-based systems in the United States, especially compared to the emissions characterization that has been done at a variety of Texas and Oklahoma feedlots. The grazing portion of the beef farm emission model is therefore less constrained and may result in the underprediction of emissions of ammonia from beef not housed on feedlots.

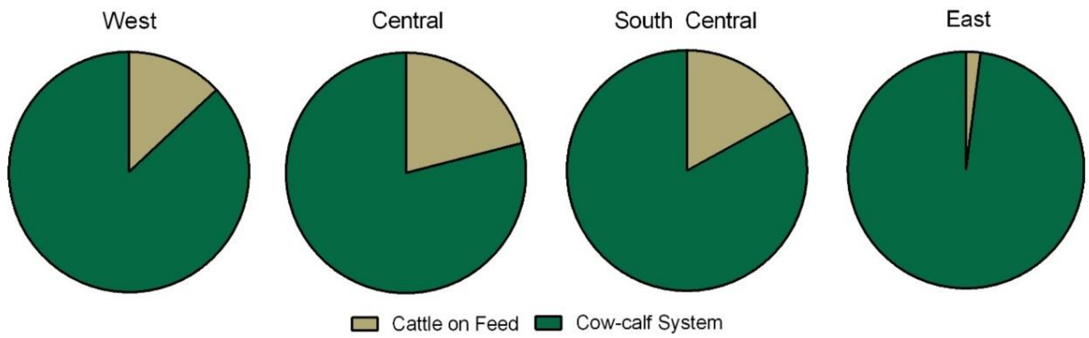


Figure 4.2. Regional distribution of beef cattle on feed. States in the West include: AZ, CA, CO, ID, MT, NV, NM, OR, UT, WA, and WY. The states in the Central region are: IL, IN, IA, KS, MI, MN, MO, NE, ND, SD, and WI. TX and OK are in the South Central region. The remaining states are in the East.

4.3.3.4 Swine Practices

There is significant regional variability in the housing types and manure management practices (in terms of storage and application) for swine production in the United States. Some of the management choices made are the result of meteorological limitations (i.e. deep-pit versus shallow-pit housing) while others are chosen for economic reasons (less expensive to use irrigation application rather than injection).

Using the information provided by NAHMS, regional distributions of management practices can be described (USDA-APHIS, 2008a, 2008b, 2008c, 2007d). The United States can be broken into three regions based on this data: the South, the Midwest, and the East. Each of these groups of states has a unique distribution of housing, storage, and application practices, seen in Figure 4.3. The recalibration of manure storage for swine production was the most significant change between the FEM between the literature evaluation in Chapter 2 and the NAEMS-based evaluation from Chapter 3, and this has resulted in a significant increase in the contribution of ammonia emissions from swine lagoon storage.

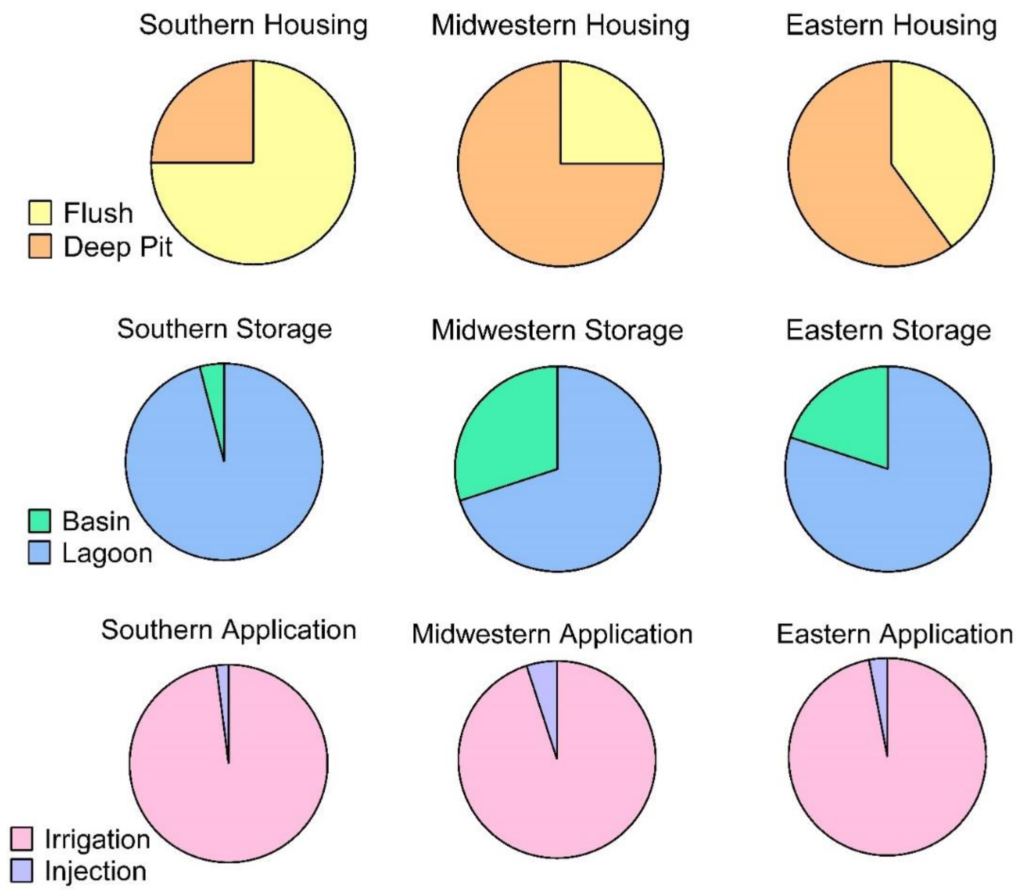


Figure 4.3: Regional Distribution of swine manure management practices. The Midwest includes: ID, IA, MN, MT, ND, NE, SD, WI and WY. The Eastern states include CT, DE, IL, IN, ME, MD, MA, MI, NH, NJ, NY, OH, PA, RI, and VT. The remainder of the states are included in the Southern region.

4.3.3.5 Layer Practices

There are two major housing types used in the production of layer chickens in the United States. These are high-rise layer houses and manure-belt layer houses. The chief difference between these two housing types is the frequency with which manure is removed; in high-rise barns, manure is removed 1-2 times each year, while manure is removed on a daily or weekly basis from manure-belt barns, which results in lower housing emissions and ammonia concentrations but leaves greater quantities in the manure that is headed toward storage and application or processing. High-rise housing operations are more prevalent than manure-belt houses throughout the United States (Figure 4.4), but manure-belt are somewhat more common in the western and central portions of the United States. There are some limitations on the abiility of the FEM for both the storage and application of poultry manure as there have been few studies to characterize these emissions. The majority of ammonia emissions from poultry are expected to be from housing (particularly for high-rise facilities).

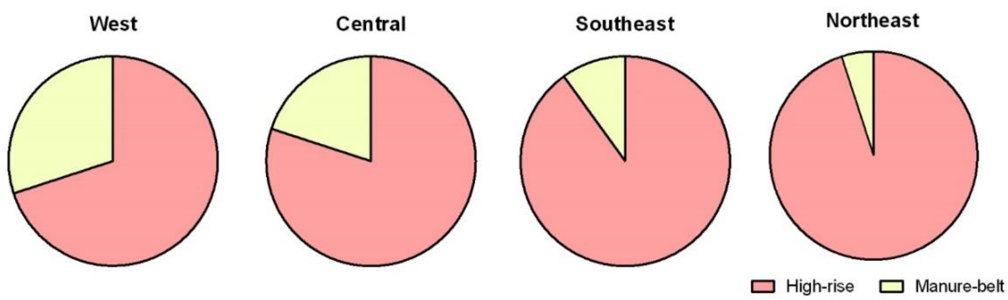


Figure 4.4. Regional distribution of layer housing types. The West includes: AZ, CA, CO, ID, MT, NV, NM, OK, OR, TX, UT, WA, and WY. The Central states are: AR, IL, IN, IA, KS, MN, MO, NE, ND, OH, SD, and WI. Southeastern states are: AL, FL, GA, KY, LA, MS, NC, SC, TN, VA, WV. The remaining states are considered to be in the Northeast.

Additionally, the most recent NAHMS information does not capture the more recent trend towards cage-free housing or pasture-raised layer chickens (USDA-APHIS, 2014a, 2014b, 2000). Cage-free housing is a relatively minor housing practice currently (<10% of all layer chickens are raised on cage free farms, but state-specific data is unavailable so this may vary significantly by state, and this may not represent a similar fraction of total eggs produced), but is poised to grow as a result of concerns about animal health and welfare and the demand for cage-free eggs increases. According to the most recently completed NAHMS, cage-free production occurs at approximately 3% of large layer operations (more than 100,000 layers), and approximately one-quarter of smaller farms. The data provided by NAHMS does not specify the fractions of total layer populations raised at particular farm sizes, but large farms have become increasingly common and it is expected that most eggs are produced from larger farms (USDA, 2014). Cage-free and organic products are more likely to come from smaller farms whose emissions have not been well-characterized in the literature. Cage-free production is more common in Europe than the United States, so emissions studies from Europe could be used to better characterize cage-free housing emissions (Charles, 2016; Shepherd et al., 2015; Zhao et al., 2015).

4.3.3.6 Broiler Practices

The major differences in broiler chicken production occur not in terms of farm type, but in the frequency with which barns are entirely cleaned out of their litter material; literature suggests that barns that are cleaned out more frequently have lower emissions than those in which litter material is built up and reused (USDA-APHIS, 2011, 2005a, 2005b, 2005c). Additional factors that may alter the emissions from these facilities include what the bedding or litter material is made up of as well as how long each barn stays empty between flocks. There is not sufficient data to include either bedding material or the time between flocks within the emissions inventory. In fact, much of the variability that might be caused by these factors on a single farm will likely be averaged out as a result of short lifecycle of these birds, which take less than two months to reach market size. Additionally, we have not included pasture-raised or organic practices as they make up a very small fraction of total bird population and the emissions from these farms has not been characterized in the literature. The limited data available regarding manure storage and application from broiler housing may result in the underestimation of ammonia emissions from this animal type.

4.3.4 Inventory Construction

As described in the previous sections, the following information is required to produce an accurate, daily time-resolved ammonia emissions inventory: daily meteorology, regional distribution of management practices, and county-level animal population data. We use the FEM described in the previous chapters to produce emissions factors for each day for each county in the continental United States for each animal type. The flow of information to produce daily, location and practice specific emission factors is shown in Figure 4.5 below.

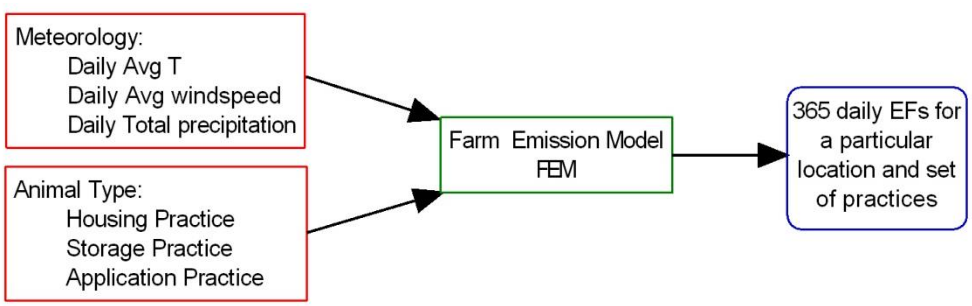


Figure 4.5. Process to produce location and practice specific daily emission factors. The farm emission model is run for all combinations of practices for each location to produce a set of daily emission factors for each location. The procedure for producing a

single emission factor for each day, weighted by the distribution of practices is shown below in Figure 4.6, where the values from 1-n describe the set of manure management practices, j specifies the day of the year, k indicates the county location, and a specifies animal type; f indicates what fraction of the animal population in a specific location is raised using a particular set of practices. The distribution of management practices is assumed to be constant throughout the year. Thus, the composite emission factor for a given day is simply the fraction of the animal population in a given location raised using a certain set of practices multiplied by the emission factor for that practice (in that location for that day) and summed over all management practices. Then, to calculate the total emissions for that animal type in that county for each day, we simply multiply by the animal population of that animal type in the county (Equation 4.2).

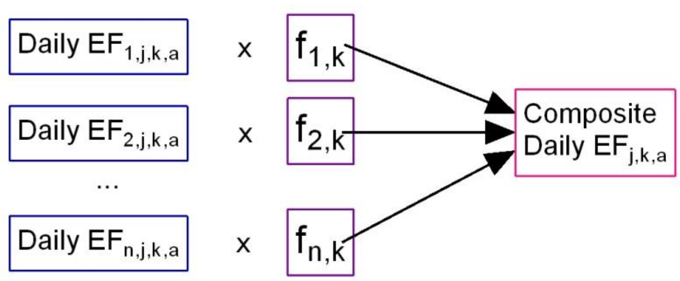


Figure 4.6. Composite emission factors for a specific day, location, and animal type.

$$Emissions_{j,k,a}\left(\frac{kg}{d \cdot county}\right) = Daily\ EF_{j,k,a} \times Population_{k,a} \qquad (4.2)$$

The total emissions in any given day may then be calculated by adding up all the emissions in each county for all animal types. This is shown in Equation 4.3. Total annual emissions for each location are calculated by summing the daily emissions over the entire year; this is described in Equation 4.4. Total annual emissions (for all animal types and all locations) are described in Equation 4.5.

$$Emissions_{j,k}\left(\frac{kg}{d\cdot county}\right)=\sum_{a=1}^{all\ animal\ types} Emissions_{j,k,a}\left(\frac{kg}{d\cdot county}\right) \tag{4.3}$$

$$Emissions_{k}\left(\frac{kg}{y}\right)=\sum_{j=1}^{365} Emissions_{j,k}\left(\frac{kg}{d\cdot county}\right) \tag{4.4}$$

$$Emissions_{total}\left(\frac{kg}{y}\right)=\sum_{k=1}^{US\ Counties} Emissions_{k}\left(\frac{kg}{d\cdot county}\right) \tag{4.5}$$

By producing the emission factors from the FEM and then weighting these results separately, it is easier to adjust the distribution of manure management practices used, which can be used to investigate how sensitive total emissions are to these practices.

4.4 Overview of Results

Results are presented in the following sections for total US livestock ammonia emissions and by animal type (swine, dairy, beef, layers, and broilers). This section will present annual fluxes of emissions (presented in terms of kg/km^2/y) for each animal type (by county) and total annual livestock ammonia flux. Daily resolved emissions of ammonia are also presented. Additional results can be found in the supplemental information for each state that shows day-to-day variability in emissions for all livestock as well as by animal type.

4.4.1 Total Annual Emissions of Ammonia from Livestock

Figure 4.7 presents a summary of the results, namely daily emissions for the year 2011 for each livestock category as well as the total livestock emissions. Annual ammonia emissions from livestock total approximately 1.7 Tg NH_3.

Based on the model results, swine contribute the most to total emissions, particularly during the summer (42%), with beef contributing 25% and dairy 12%. Layers and broilers contribute 5% and 15% respectively. The swine contribution is higher than originally expected, likely as a result of the increase in swine population over the past 10-15 years. Additionally, the swine emissions from manure storage were also increased as a result of the higher ammonia emissions that were observed during NAEMS (and to which the model was re-tuned, described in the previous chapter).

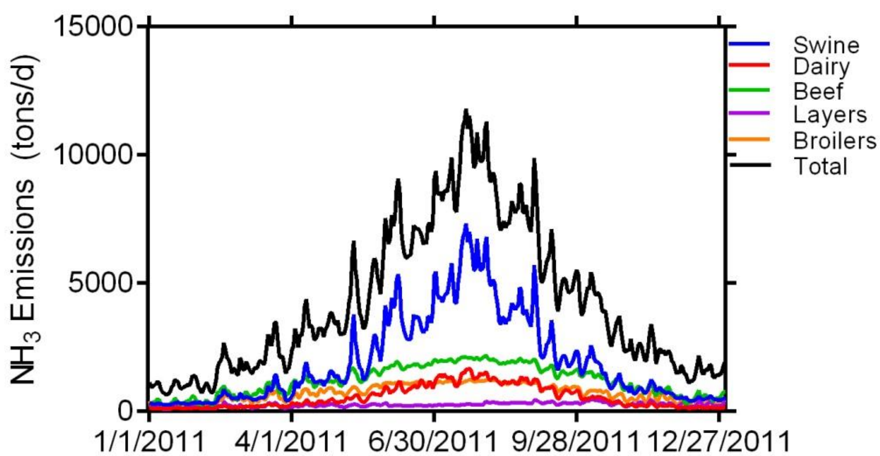


Figure 7. Total US Livestock Ammonia Emissions for swine, dairy, beef, layers, and broilers in tons NH3 emitted per day

There is an obvious seasonality to the total ammonia emissions, largely driven by the emissions from swine production in the United States. Beef emissions make comparable contributions to total emissions during the winter as swine; dairy farms have a similar seasonality to the beef emissions. Next most important to total emissions are broilers, and finally, layers make the smallest contribution to the national emissions inventory for ammonia from livestock. Furthermore, there is a weaker seasonal trend for the poultry emissions sources when compared to swine and both types of cattle.

In Figure 4.8, we compare our results to those presented by Pinder et al. (2006) by adding up daily emissions to get total livestock ammonia emissions in Gg/month. The results from the daily model are approximately 15% lower than those in the monthly inventory from Pinder.

The seasonality of overall emissions in this work is more driven by swine emissions; the previous model's seasonality is more similar to the seasonal patterns in beef and dairy cattle ammonia emissions. This means that the current inventory predicts higher summertime and lower wintertime emissions of ammonia than in the previous study. It should also be noted that the meteorology used in the two inventories is different (the Pinder inventory is based on long-term meteorological averages while the current work is based on actual daily meteorology from 2011).

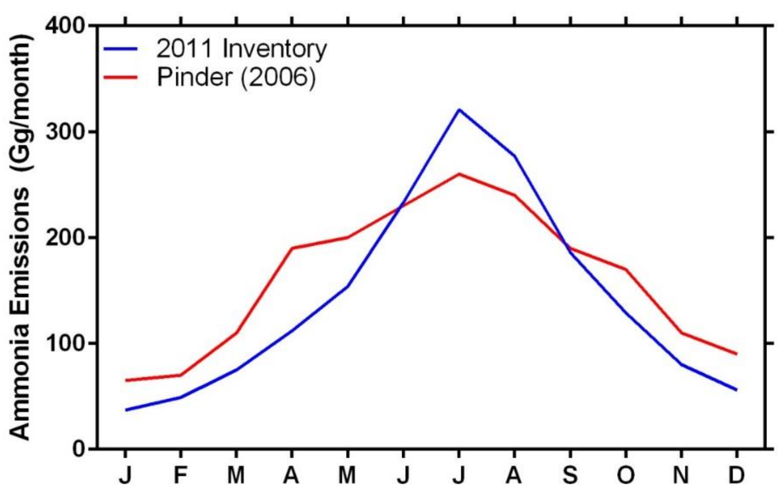


Figure 4.8.Comparison of livestock NH_3 emissions inventories from Pinder (2006) and 2011 inventory.

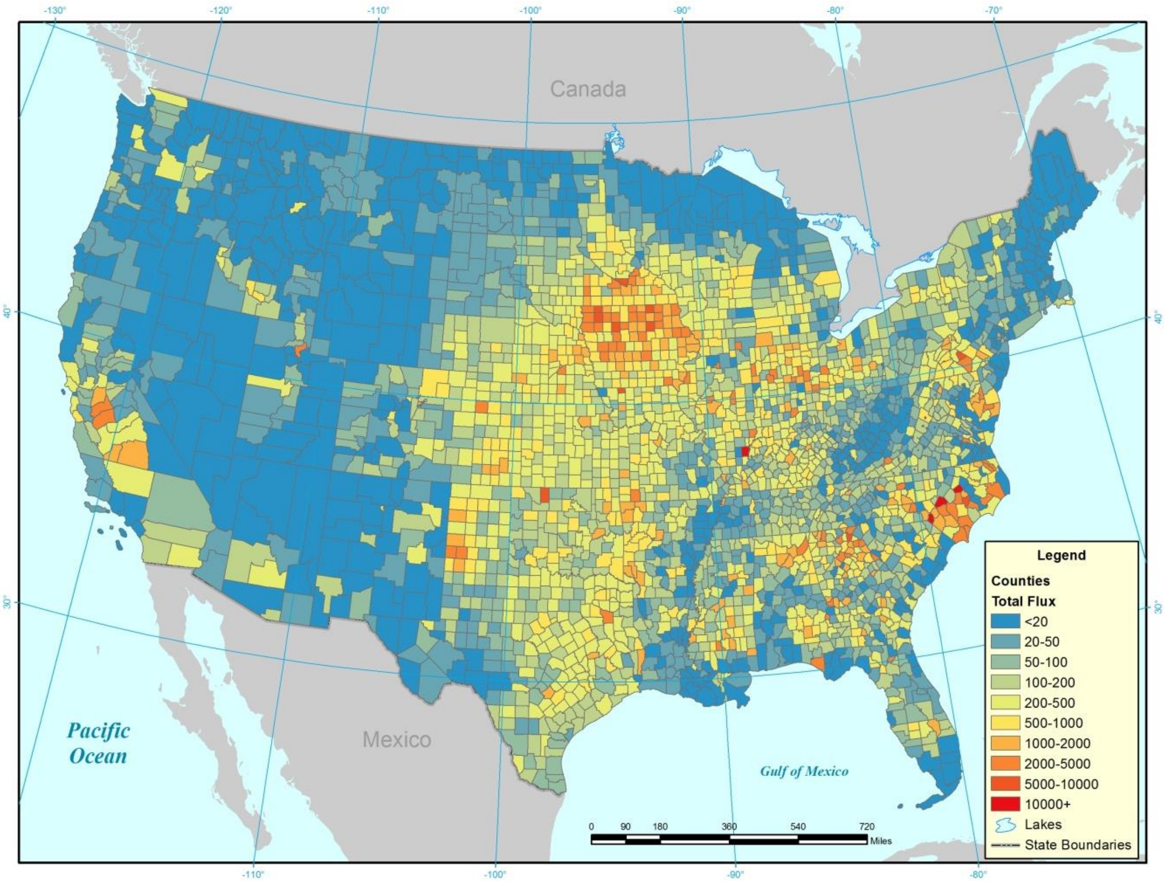


Figure 4.9: Annual Ammonia flux from all livestock types (swine, dairy and beef cattle, and layer and broiler chickens) for each county in the United States (kg/km^2/year).

The map in Figure 4.9 shows annual-average ammonia emissions fluxes by county across the United States. In this figure, areas with intensive agriculture stand out as expected. For example, the San Joaquin Valley of California is evident (home to 5 million cattle and 60 million chickens), as is the state of Iowa (home to more than 20 million swine, 54 million chickens, and 4 million cattle), and the Southern Coastal plain of North Carolina (with 9 million swine and 63 million chickens).

4.4.2 Swine Emissions Inventory

The US swine population is located primarily in the Midwest and Southeast. Figure 4.10 shows a typical breakdown of where in the manure management process ammonia emissions occur. In the Midwest, where deep-pit housing dominates, there is a roughly even split between housing emissions, storage emissions, and application emissions. In the Southeast, shallow-pit housing dominates (shallow-pit houses are cleaned more frequently than deep pit houses, and so the manure removed from them is richer in nitrogen), and so emissions from manure storage, mostly (anaerobic) lagoons dominate, contributing more than half the total emissions from a farm.

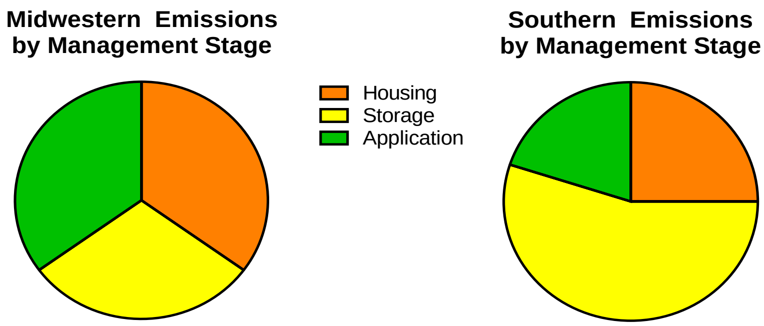


Figure 4.10: Typical regional contributions of housing, storage and application to total swine emissions.

However, the breakdown between management stages is not consistent throughout the year—during the winter, housing emissions contribute more to total emissions while storage and application emissions approach zero. This variation results from the fact that houses must be maintained at a comfortable temperature while manure storages may freeze and application may not be occurring during the winter. Unfortunately, the frequency of manure application has not been well described in the literature, in NAEMS or by NAHMs, so we simply assumed that manure was applied daily. This may result in the overestimation of emissions from application during the summer and winter and underestimation in the spring and fall when manure is known to be applied as fertilizer.

Figure 4.11 shows the daily variability in ammonia emissions from the model as well as emissions that are 125% of the model's estimate and 75% of the model results. These values were selected as they represent a conservative estimate (on the lower end of expected uncertainty) of the uncertainty inherent in our model resulting both from measurement uncertainties (on which model evaluation was based and was described in Chapters 2 and 3) and from uncertainties in the distribution of practices throughout the country as described by the regional distributions in practices from NAHMS.

For comparison, emissions recommendations from previous studies by Faulkner and Shaw (2008) and Battye et al. (1994, 2003) are also presented. These represent levels of emissions currently in use within inventories.

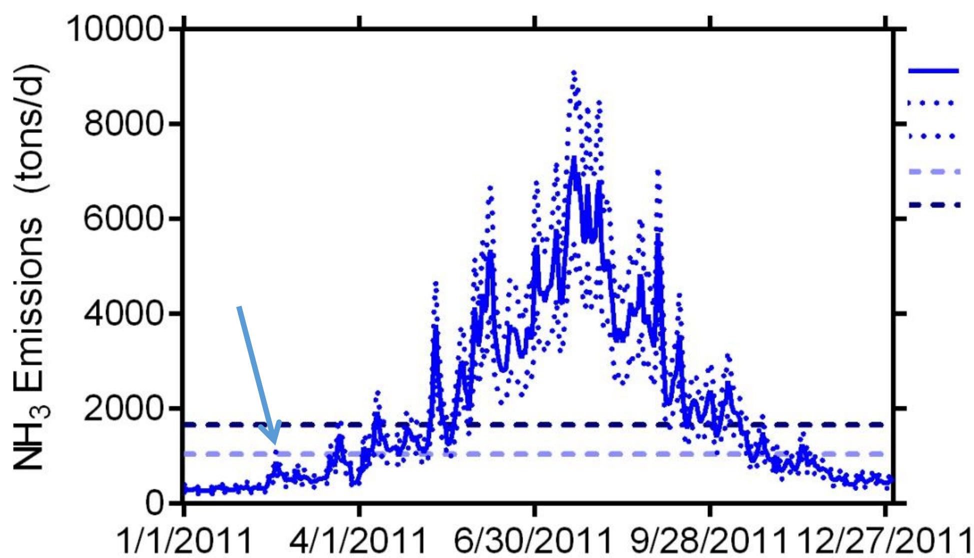


Figure 4.11. US Swine Emissions (2011) presented in tons NH_3/d. The arrow in the figure indicates a particularly warm day (for the winter) in both North Carolina and Iowa.

Wintertime emissions are lower than the constant emissions recommendations of Battye and Faulkner, but summertime emissions are much higher. The higher emissions from the farm emission model result from higher expected emissions from manure storage, particularly during warm temperatures, especially in the Southeast. These high manure storage emissions were first seen at the farms studied during NAEMS.

US emissions from swine are located mostly in the Midwest (Iowa, Indiana, and Illinois) and Eastern North Carolina. This is shown in Figure 4.12 in terms of annual total ammonia flux per square kilometer. Even though there are half as many swine in North Carolina as Iowa, the warmer climate drives higher emissions from each animal.

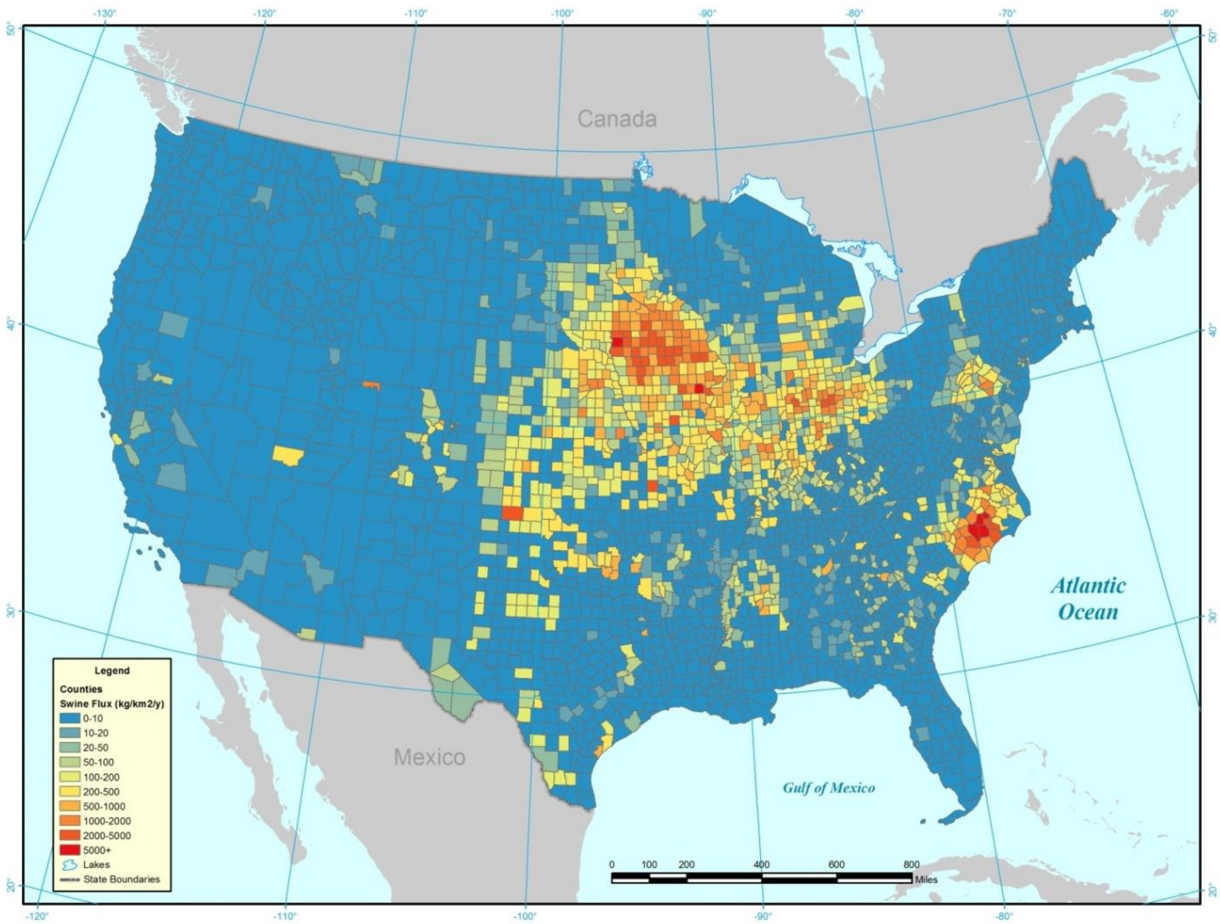


Figure 4.12. Annual swine ammonia flux (normalized by county area) in $kg/km^{2/}y$. Intense production regions in Iowa and North Carolina are noted.

The magnitude of total swine emissions can vary by an order of magnitude (or more) between a cold winter's day and a warm summer one. Emissions can also vary dramatically between one day and the next. For example, as noted by the arrow in Figure 11, the emissions on February 16 were 240 tons lower than on February 17; this represents a more than 40% increase in total emissions from one day to the next. February 17 was significantly warmer than February 16 in both North Carolina and in Iowa, home to nearly half of the country's swine population. Variability in total emissions is even more obvious during the summer when emissions are at a higher level in general. The model's ability to show the day-to-day variability in ammonia emissions, as evident in the model results, will be particularly helpful in identifying the potential for air quality events that occur during winter where a single warm day can produce a "burst" of emissions resulting in massive PM formation and causing relatively rural locations (like most of Iowa) to be in non-attainment for the fine particulate matter ambient air quality standard (Stanier et al., 2012).

In addition to the day-to-day and seasonal variability in emissions that are captured by this approach to inventory construction, we can also identify regional differences in emission factors for swine. This is shown in Figure 4.13.

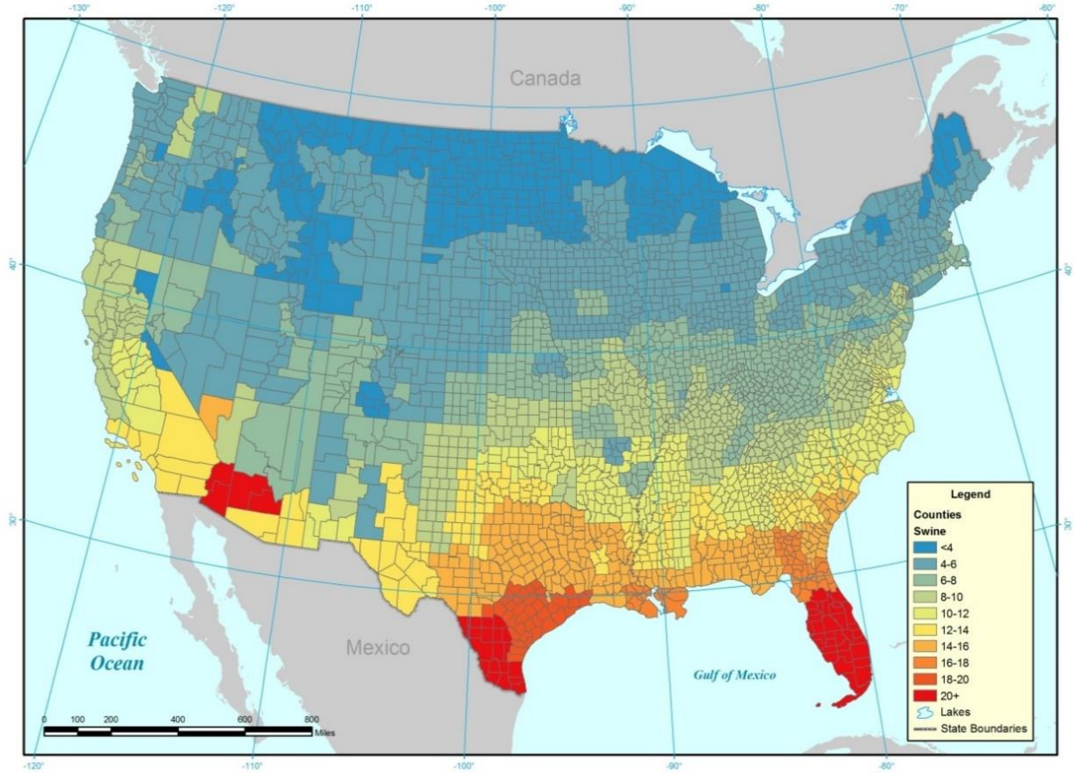


Figure 4.13. Regional distribution of swine annual emission factors (shown in kg/animal/year)

4.4.3 Beef Emissions Inventory

The US beef inventory is more dispersed throughout the country than either swine or poultry populations. Most beef cattle are raised in the Central United States, with a greater fraction of these animals on feedlots here than in the rest of the country. Emissions rates from feedlots are much greater than from cattle on pastures for a comparable number of animals because of the inability of waste (and the nitrogen in the waste) to infiltrate the surface of the feedlot. As stated previously, there have been few studies which monitor ammonia emissions from cow-calf or pasture based systems. The emissions from grazing animals are expected to be significantly lower than those from animals housed on feedlots, but results range from 2 kg/animal/y – 9 kg/animal/y.

Even though emissions from grazed animals are much lower (10-15% of the emissions relative to an animal housed under the same meteorological conditions on a feedlot), because so many fewer animals are on feed, both grazed and fed beef cattle are important to the overall national inventory of emissions. Daily-resolved beef ammonia emissions are shown in Figure 4.14.

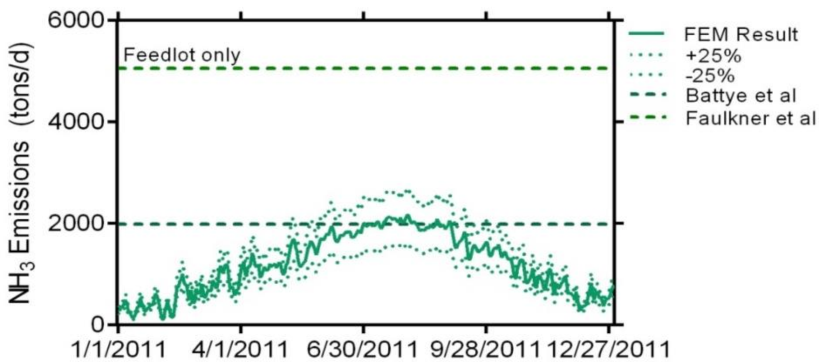


Figure 4.14. US Beef Emissions (2011) presented in tons NH_3/d.

The regional distribution of annual ammonia fluxes are shown in Figure 4.15.

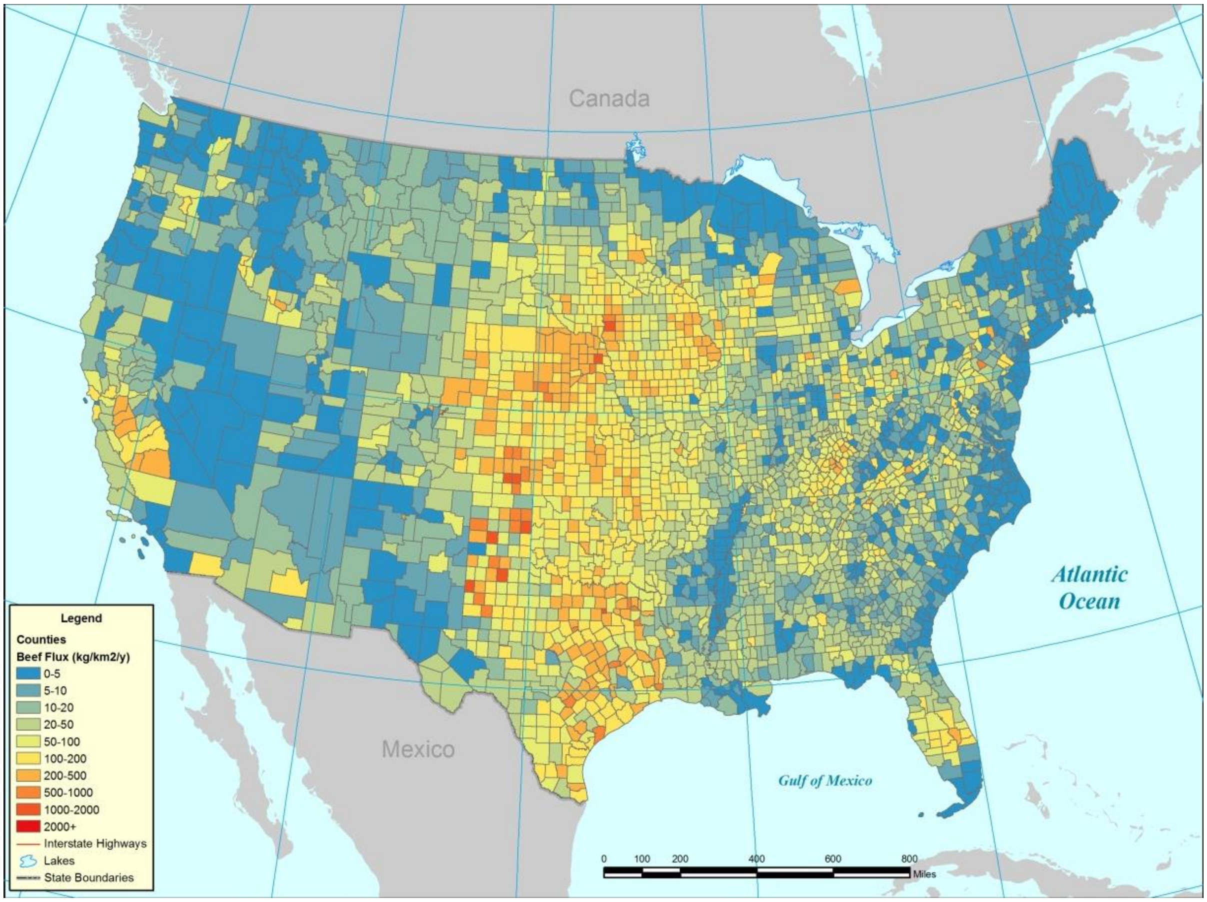


Figure 4.15. Annual beef ammonia flux (normalized by county area) in kg/km$^{2/}$y.

Annual ammonia fluxes from beef are greatest in the Great Plains region of the country, particularly through Texas, Oklahoma, Kansas and South Dakota.

Using the FEM also allows us to visualize regional differences in annual emissions factors. For beef, this is driven not only by the local meteorology, but also by the fraction of animals in a particular location that are raised on feed (as opposed to pasture). In locations where the fed beef fraction is particularly high, emissions are higher than in some locations which may have warmer temperatures but a smaller fraction of cattle raised on feed. This is shown in Figure 4.16.

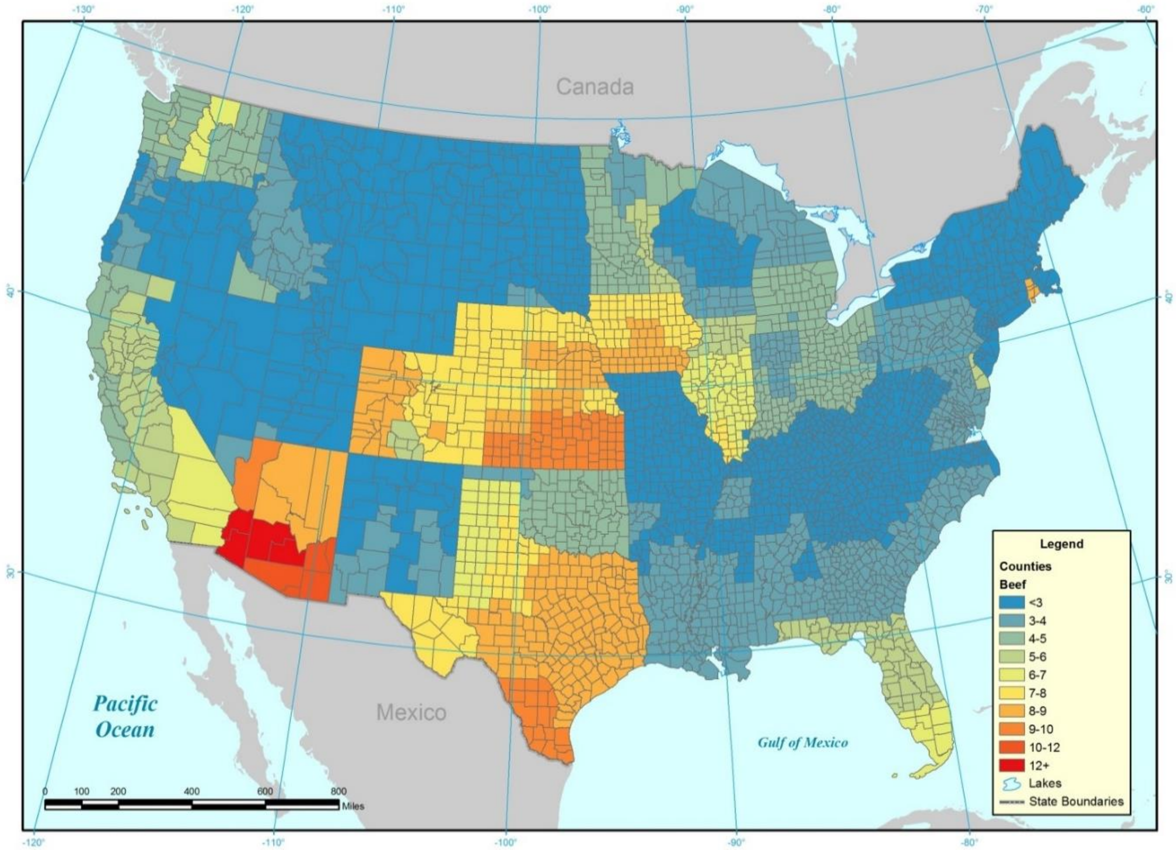


Figure 4.16. Regional distribution of beef annual emission factors (shown in kg/animal/year)

4.4.4 Dairy Emissions Inventory

Dairy production in the United States is primarily located in the Northeast (New York and Pennsylvania), the Upper Midwest (especially Wisconsin), and the San Joaquin Valley of California. The results for the dairy inventory are expected to be very similar in seasonal distribution to those produced previously by Pinder et al. (2004); however, the animal populations have been updated and practices have been altered to reflect the distribution of manure management practices presented in the most recent NAHMS reports. Annual dairy emissions range from 10-60 kg/animal, with significant emissions from each part of the manure management process. As with swine, we assumed daily manure application because there was not enough information to assume any other frequency; this may cause over-predictions in winter and summer emissions from application and under-predictions in the spring and fall. Daily variability in dairy ammonia emissions is shown in Figure 4.17. Again, comparisons to recommendations from previous studies are shown for context.

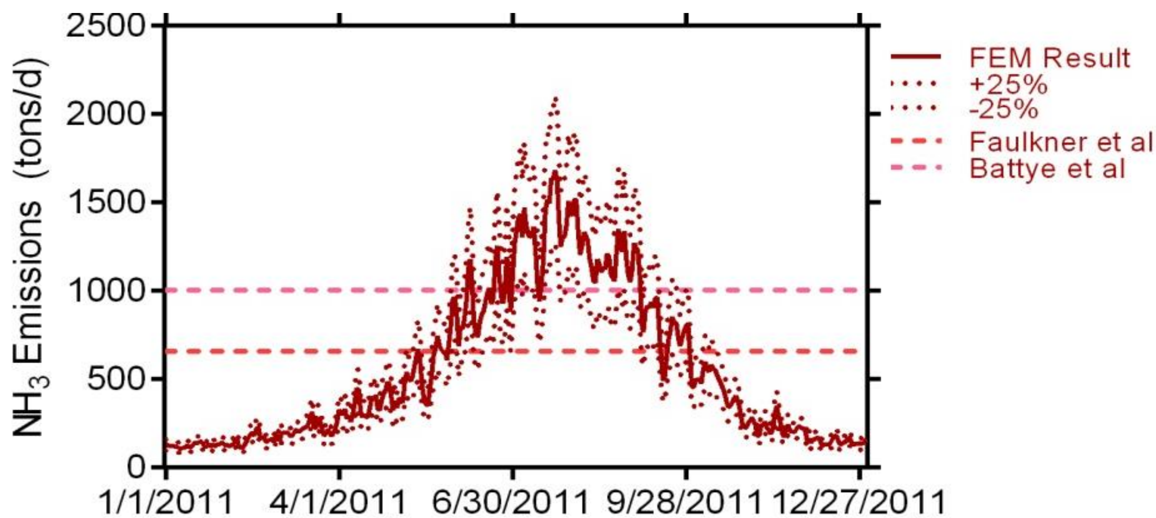


Figure 4.17. US Dairy Emissions (2011) presented in tons NH_3/d.

The dairy population distribution is evident in the map of annual ammonia fluxes from dairy cattle. This is shown in Figure 4.18. Additionally, by using the FEM, we were able to determine the regional distribution of emissions factors for dairy production in the United States. Here, we have the annual emission factors (per animal), as driven by both meteorology and the regional distribution of practices.

Based on this assessment, it appears that the model may be under-predicting the emissions associated with farms located in colder regions. Furthermore, due to the limited information regarding dairy practices that was available from the recent NAHMS report, the distribution of manure management practices may (provided in % of operations rather than animal population) be giving to much weight to practices used on smaller farms. This is shown in Figure 4.19.

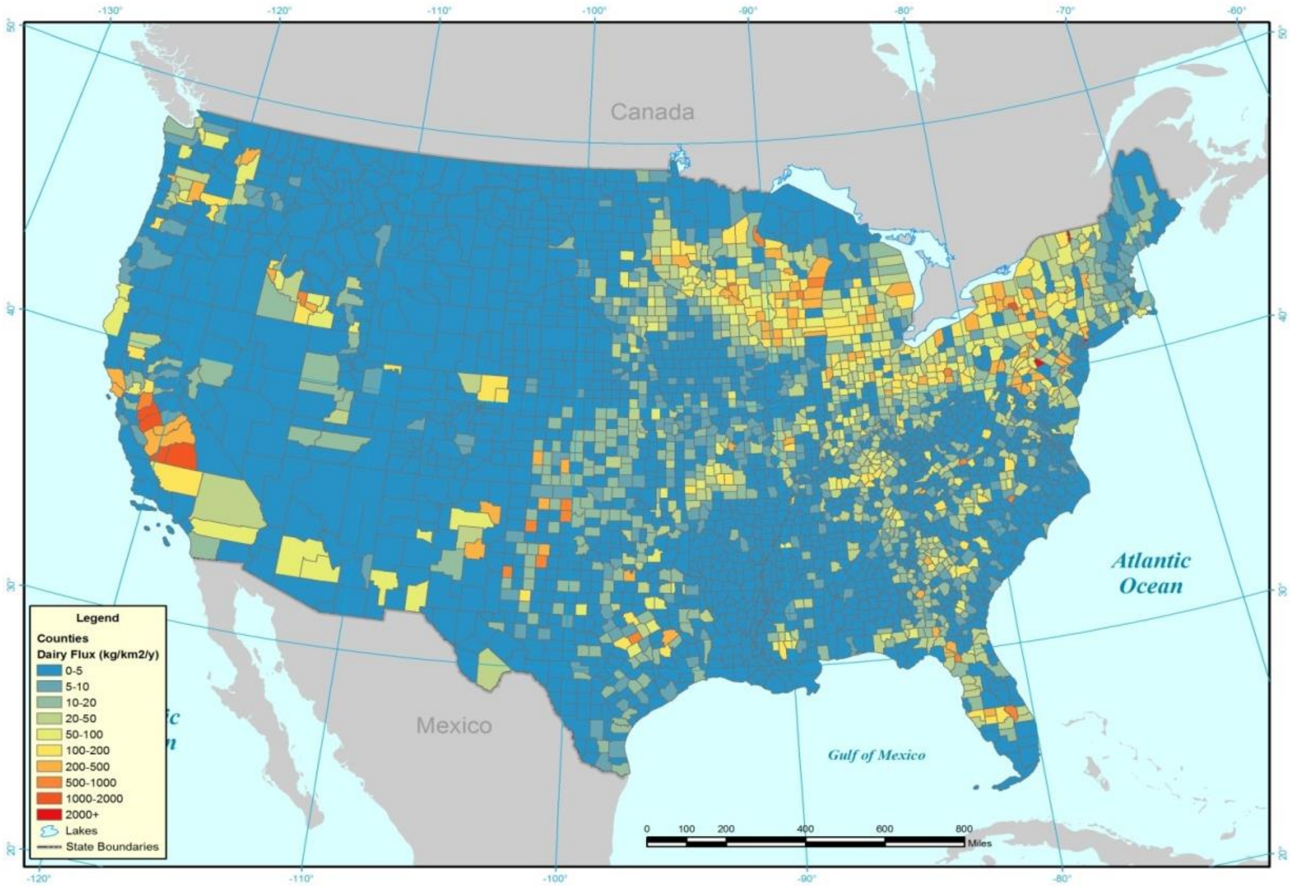


Figure 4.18. Annual ammonia flux (normalized by county area) in kg/km$^{2/}$y. Dairy population is most concentrated in the San Joaquin Valley, CA and Wisconsin, Pennsylvania, and New York.

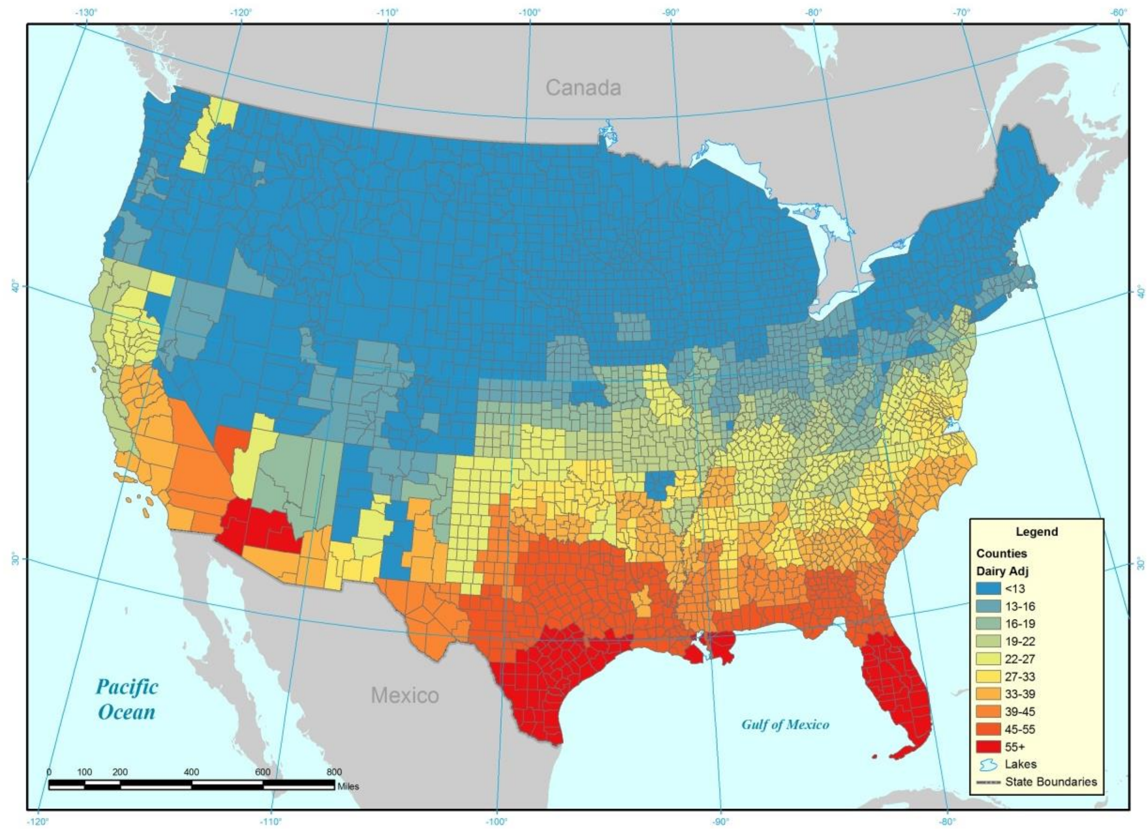


Figure 4.19. Regional distribution of dairy annual emission factors (shown in kg/animal/year)

4.4.5 Layer Emissions Inventory

Poultry ammonia emissions have not been well-described in the literature, especially for manure storage and application. As such, it is likely that our overall emissions may be an underestimate. Emissions from poultry in general are less driven by meteorological conditions than other animal types. There are also interesting trends in layer ammonia emissions (with respect to temperature) that were identified during the evaluation of the FEM with data from NAEMS in the previous chapter—wintertime emissions tended to be higher than summertime emissions, but within a single warm season month, warmer temperatures were associated with higher daily emissions. In this inventory, we have attempted to capture both scales of temporal variability in ammonia emissions.

Day-to-day variability in ammonia emissions from layers is presented in Figure 4.20 below. Previous emissions recommendations are also shown for context.

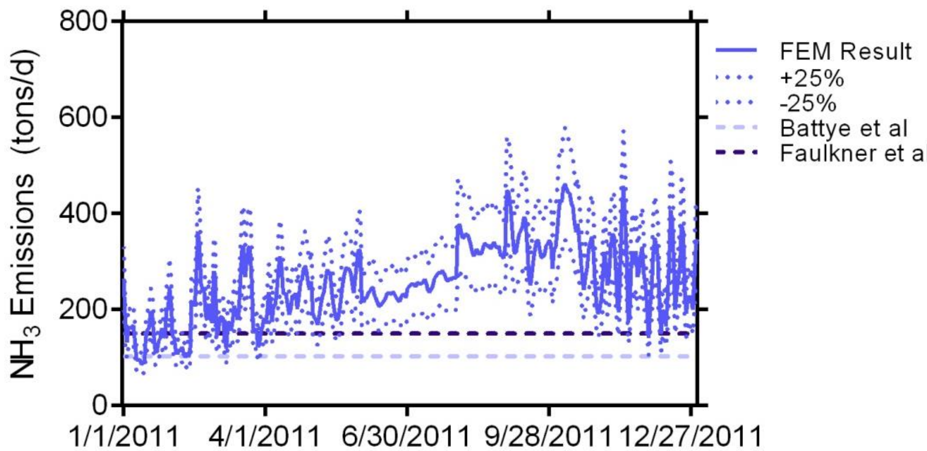


Figure 4.20. US Layer Emissions (2011) presented in tons NH_3/d.

As expected the emissions are less driven by temperature than seen for swine, dairy cattle and beef cattle. Furthermore, it seems that the results from the FEM are slightly higher than previous recommendations for layer emissions.

Layer production is particularly concentrated in a handful of counties, not necessarily across entire states. Particularly intense emissions fluxes are shown in counties in the Midwest (Iowa, Illinois and Indiana) and throughout the Southeast, from Texas to North Carolina. These results are shown in Figure 4.21.

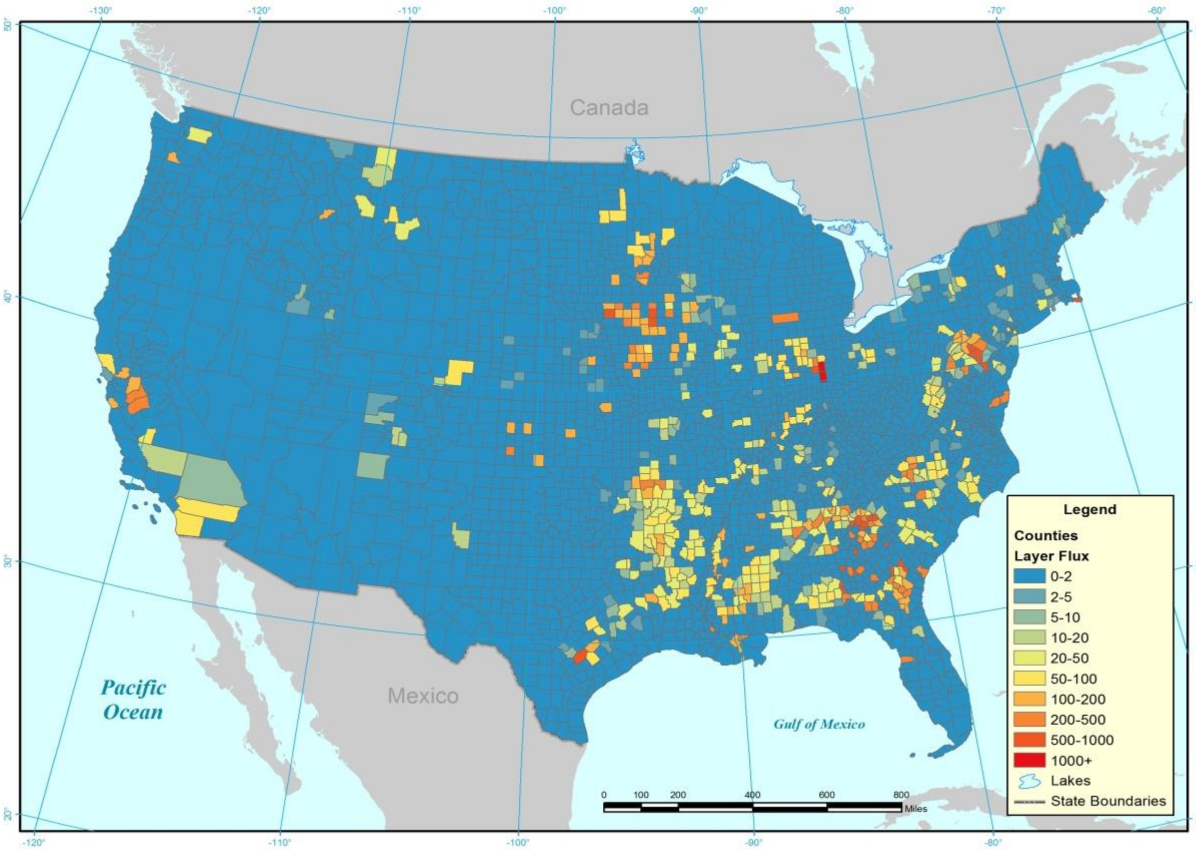


Figure 4.21. Annual ammonia flux (normalized by county area) in kg/km$^{2/}$y. Layer population is most concentrated in counties throughout the Midwest and across the Southeast.

The regional distribution of annual emission factors is shown in Figure 4.22 below; this highlights the model's ability to capture not only temporal variability in emissions but also spatial variability. The differences in ammonia emissions factors result from both differences in meteorology as well as differences in the distribution of manure management practices, especially the prevalence of high-rise versus manure-belt housing.

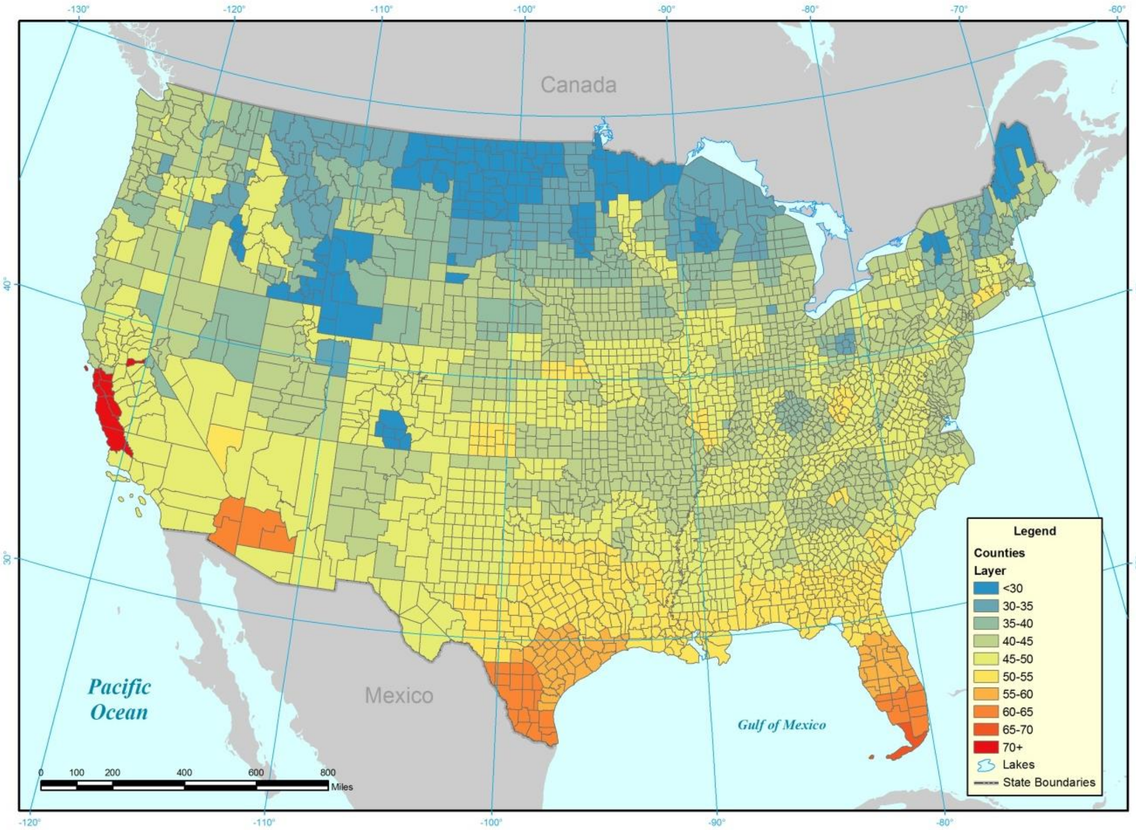


Figure 4.22. Regional distribution of layer annual emission factors (shown in kg/animal unit/year)

Overall, layer chicken emissions are a minor contributor to the national emissions inventory, but for some states, highlighted in Figure 21, layer emissions can be very important. For example, layer emissions make up approximately 20% of annual emissions in Alabama and Florida, 40% of emissions in Maine, and nearly 50% of emissions in New Jersey.

4.4.6 Broiler Emissions Inventory

Similar to layer emissions, broiler emissions are less temperature driven than emissions from cattle or swine. Additionally, the fact that the lifecycle of broiler chickens is so short (6-8 weeks) makes emissions characterizations difficult. For the purposes of this inventory, we assumed a single age for the chicken population for the model since any age variability is likely to be averaged out at the resolution at the farm-level and the level for which this inventory is created (county resolution). Emissions from broiler litter storage and application or disposal have not been well-characterized in the literature, and it is expected that the emissions inventory as presented may underestimate broiler emissions from manure storage and application.

Total US broiler ammonia emissions with daily resolution are shown in Figure 4.23. Recommendations from Faulkner and Battye are shown as a point of comparison to our daily variable emissions inventory.

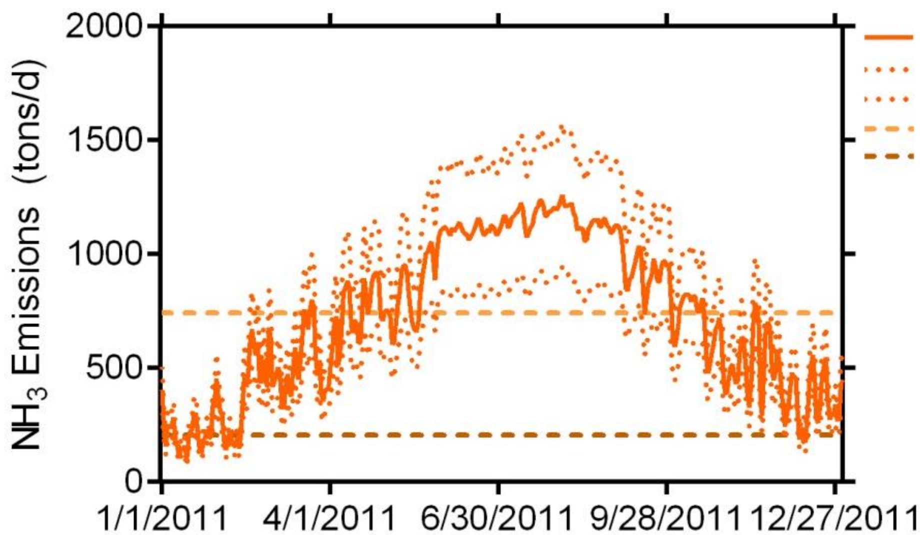


Figure 4.23. US Broiler Emissions (2011) presented in tons NH_3/d.

Broiler production is largely concentrated in the Southeastern and Mid-Atlantic states. As the chickens (particularly when they are small) need to be kept warm, it is likely easier to maintain the optimal environment in these regions. The annual broiler ammonia flux (normalized to county area) is shown in Figure 4.24.

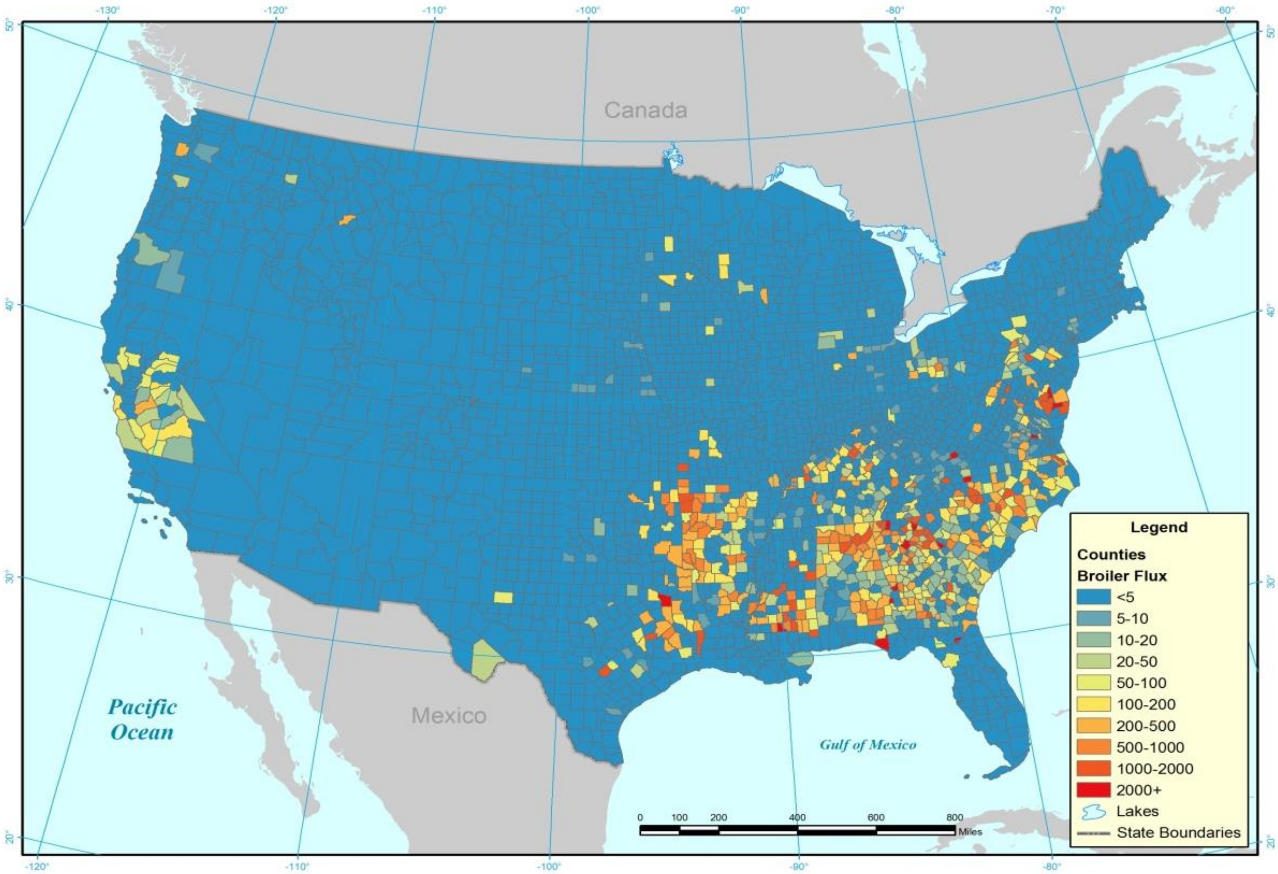


Figure 4.24. Annual broiler ammonia flux (normalized by county area) in kg/km$^{2/}$y. Broiler population is most concentrated in counties throughout the Mid-Atlantic and across the Southeast.

The regional distribution of annual emission factors for broilers, highlighting the model's ability to differentiate spatial variability in emissions is shown in Figure 4.25. Warmer temperatures are clearly associated with higher annual emission factors than cooler ones for broilers; differences in meteorology drive the spatial and temporal variability in emissions from broilers in our inventory.

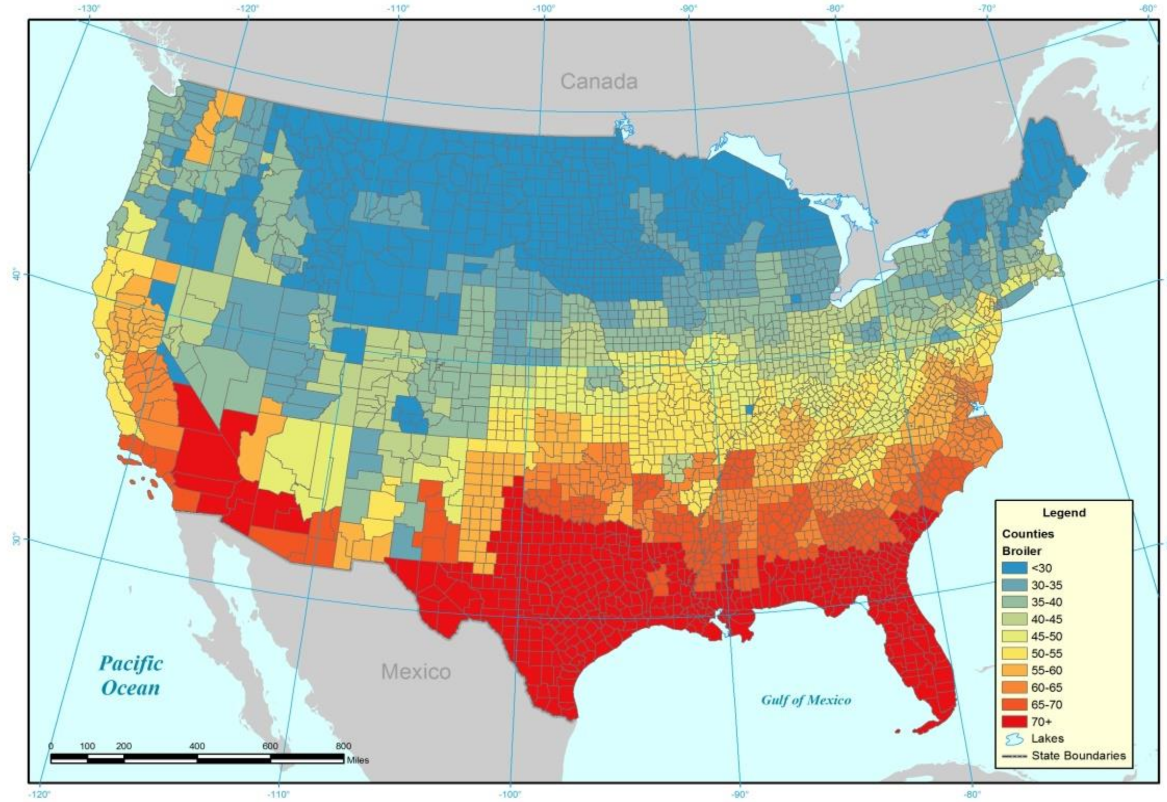


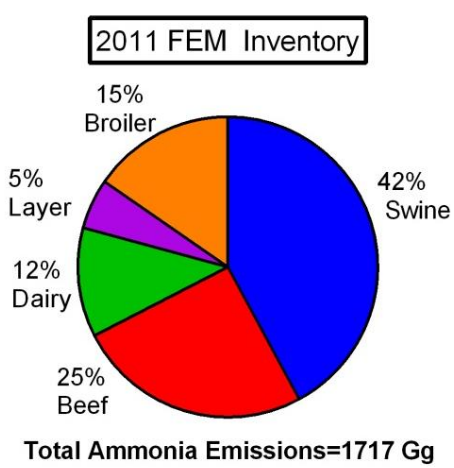
Figure 4.25. Regional emission factor distribution for broilers, in kg/animal unit/year.

5 Discussion

In this chapter, we developed a county-level, daily-resolved ammonia emissions inventory for swine, beef cattle, dairy cattle, layers and broilers across the United States. Swine production contributes approximately 42% of the total ammonia emissions, with beef contributing 25%, broilers contributing 15%, dairy cattle contributing 12% and layers represent the remaining 5% of total annual livestock emissions.

The swine emissions from our model are higher than previous studies as a result of new data from NAEMS indicating that very high summer manure storage emissions are possible. It is also possible that beef emissions are under-predicted; grazing cattle emissions are not well-characterized so our parameterization of this management practice is based on relatively little data, but these emissions are very important. Emissions from poultry are less seasonally variable than other animal types, but production is more concentrated in smaller regions, meaning that they can be very locally important.

In total, US livestock emissions in 2011 were approximately 1.7 Tg according to our model. Of this total, 723 Gg are from swine, 435 Gg are from beef, 202 Gg are from dairy, 93 Gg are from layer chickens, and 260 Gg are from broilers. The total ammonia emissions from swine, beef, dairy, layer and broiler production from the 2011 NEI were 1.97 Tg with contributions of 435 Gg from swine, 534 Gg from beef, 535 Gg from dairy, 146 Gg from layers, and 319 Gg from broilers (USEPA-OAQPS, 2015a, 2015c, 2013). This means that our model predicts significantly greater emissions from the US swine population than the NEI, while our model's prediction of dairy emissions is 333 Gg lower than the NEI estimate. Beef, layer and broiler emissions are all also under predicted but there is only a 20-30% difference in the inventories for these animal types all shown in Figure 4.26 below.



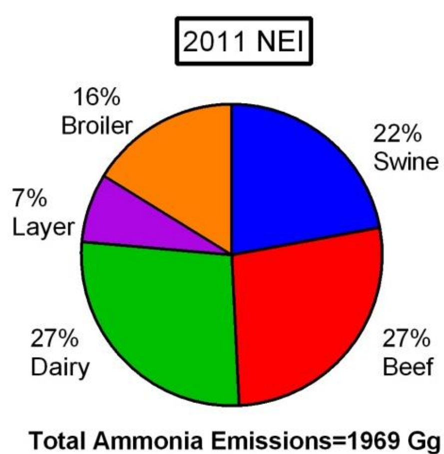


Figure 4.26. Comparison of NEI 2011v2 results and our FEM-based inventory.

Our 2011 ammonia inventory is significantly lower as a result of a number of factors. First, we expect our beef inventory to possibly be underestimated due to the limited characterization of grazing beef emissions in the literature. The spring and fall manure application from swine and dairy may also be underestimated due to the limited information available for the timing of manure application throughout the country. It is not clear what is driving such a large difference between the ammonia emissions from dairy cattle in our inventory and the 2011 NEI.

The annual emission factors (sum of daily emissions factors) from the dairy FEM are generally in line with the results reported for the seasonal model developed by Pinder et al. (2004b) which suggested that annual per animal emissions from dairy production were between 13-55 kg/animal/year. After further investigation, some locations with particularly cold climates may be under-predicted currently, as the initial model was unlikely to have been evaluated for performance at such low temperatures as are observed on a daily basis. The previous tuning of the model was done with monthly average temperatures, and these monthly average temperatures tend to be less extreme in terms of both highs and lows. There were also a few locations which had higher than 55 kg/animal/year emission factors and this can be explained also by the lack of characterization of dairy emissions at these extreme temperatures. Even considering that the emissions from dairy from the FEM may be low, that would likely still be insufficient to explain the fact that the FEM predicts 2.5 times lower dairy emissions than the NEI.

CHAPTER 5: CONCLUSIONS

The main objectives of this work were to do the following:

- Develop farm emissions models (FEMs) for beef cattle, swine, broiler and layer chickens based on previous research (Pinder et al., 2007, 2006, 2004a, 2004b) on dairy cattle
- Evaluate these new FEMs using observations from the existing literature and the National Air Emissions Monitoring Study in their ability to capture seasonal and day-to-day variability as well as their ability to capture the differences between different manure management practices
- Construct daily-resolved, county-level ammonia emissions inventories for beef cattle, swine and poultry, using meteorology from 2011
- Compare these inventories to the most recent National Emission Inventory, NEI2011v2

The work began with the modification of an existing semi-empirical process-based model framework for the characterization of ammonia emissions from a variety of livestock types and for a range of manure management practices. The evaluation of these FEMs against literature data helped us identify gaps in knowledge, including the limited characterization of ammonia emissions from pasture-raised cattle and the storage and application of poultry manure, as well as the inherent difficulties associated with the measurement of ammonia emissions from open sources, such as storage lagoons, commonly used in the production of swine and dairy cattle in particular.

Based on the literature evaluation, we determined that the model was able to capture 20%-70% of the variability in observed emissions.

Further evaluation of the model was completed using data from the National Air Emissions Monitoring Study (NAEMS) campaign. As a result of the higher time resolution of this data, we were able to further test and improve the farm emission model to capture day to day variability in ammonia emissions; seasonal model-measurement correlations varied from farm to farm and animal type, but were comparable to the evaluations based on literature, mean fractional biases typically less than 20% and mean fractional errors generally in the range of 20-40%. The model was slightly less skilled in accurately capturing the day-to-day variability in ammonia emissions, but for half of the farms evaluated, the daily correlation coefficient was greater than 0.25, demonstrating that a component of the daily variability can be explained simply with daily variation in meteorology, for all but one of the remaining farms, the model had little skill (-0.04<R-value<0.24). One dairy lagoon performed particularly poorly the correlation coefficient is -0.76. The farms for which model-measurement agreement is poor results partly from the difficulty of performing these measurements, accounting for dispersion, and inferring emissions, and partly from factors that may not be included in the FEM.

The next objective of this work was to use the farm emissions models developed and evaluated using the literature and NAEMS data to construct a highly-resolved ammonia emission inventory for beef cattle, swine, broiler chickens, and layer chickens, as well as update the earlier dairy inventory from Pinder et al. This daily-resolved inventory used daily meteorology (wind speed, average temperature, and precipitation) from the National Climate Data center for 2011. Additionally, the inventory used regionally specific inputs for the distribution of practices used in manure management; inventory accuracy could likely be improved if data about practices was available with greater spatial resolution, and if they were available in terms animal populations raised with certain management practices, not just in terms of farming operations. Another key feature in our inventory is that we produce daily composite emission factors for each county and then multiply by the animal population separately from the model run. Again, if more highly time-resolved data for animal population was available, we might be able to further reduce the uncertainties in our ammonia emissions inventory. Annual total emissions from our inventory were 1.7 Tg for 2011, with 42% of the ammonia emissions from swine production, followed by 25% from beef cattle, 15% from broiler production, 13% from dairy cattle, and 5% from layer production.

An emissions inventory is most valuable if it accurately reflects the actual emissions from a particular source. Following the construction of the county-level daily resolved beef, dairy, swine and poultry inventories, we wanted to compare the total swine, beef, dairy, layer and broiler emissions to those reported in the 2011 NEIv2. The 2011 NEI has a total of 1.9 Tg of emissions from these types of animals, which is roughly 200 Gg greater than the FEM based inventory. The largest differences between the two overall inventories are for swine (our inventory predicts 280 Gg greater emissions) and for dairy cattle (the NEI estimates 300 Gg more ammonia than our inventory). More detailed evaluation of the inventory will need to be completed in order to ensure it accurately captures the spatial and temporal variability in emissions across the United States. This will involve additional comparisons at a variety of spatial and temporal scales of our model's inventory to the 2011 National Emission Inventory. Additionally, we will need to determine whether the emissions inventory produced by the FEM approach results in results in accurate ambient ammonia and ammonium concentrations via the use of a chemical transport model and comparing to observations from the Ammonia Monitoring Network (AMoN) and the National Atmospheric Deposition Program (NADP). Furthermore, we will investigate the uncertainties in our model-based inventory as it relates to sources of variability not captured by the model, data limitations (especially with respect to the distribution of management practices throughout the United States), and error and uncertainty in the emission factor measurements on which the FEM-based approach relies.

The daily emission inventory produced with our FEM-based approach offers a valuable contribution in capturing the spatial and temporal variability in ammonia emissions from swine, dairy, beef, and poultry production in the United States. However, the model is only as accurate as its inputs, and there are many details in the model that would benefit from additional observations.

Ammonia emissions measurements of pasture or range-raised cattle, layer and broiler manure storage, and poultry manure application would all be particularly helpful to more accurately constrain the model. Additionally, better reporting of the conditions under which manure ammonia emissions measurements were completed, in terms of both meteorology, manure and feed characteristics, and the methods used to make the ammonia emissions measurements would help more accurately constrain the model's inputs and improve its performance. In the course of inventory development, there was only regionally-specific data available to describe the manure management practices used at farms across the country. More detailed practice information would reduce the number of assumptions required to compute the emissions inventory.

The ability to model the spatial and temporal variability in ammonia emissions in the United States is an important step towards understanding the impact of livestock production on particulate matter formation and local and regional air quality. In this work, we have developed and evaluated farm-level models, using data from the literature and the National Air Emissions Monitoring Study, to characterize daily ammonia emissions variability from swine, beef, dairy, layer, and broiler production in the United States. Additionally, we have constructed a county-level daily emissions inventory, and compared its results to those reported in the 2011 National Emission Inventory. Future work still needs to be done to better constrain ammonia emissions from poultry manure storage and application and cattle raised on pasture through additional measurement campaigns, and further evaluation of the FEM-based emissions inventory needs to be done.

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SUPPLEMENTAL INFORMATION

SUPPLEMENTAL TABLES

Table S2.1. Sources of data used for model tuning and evaluation for beef, swine, and poultry

Animal	Submodel	Data sources for tuning	Data sources for evaluation
Beef	Housing (feedlot)	1	1, 2
	Application	11	11
Swine	Housing	3	3, 4
	Storage	9	9, 10
	Application	12	12
Broilers	Housing	5	5, 6
	Application	13	13
Layers	Housing	7	7, 8
	Application	14	14

1. (Cole and Defoor, 2006; Hristov et al., 2011; Todd et al., 2008, 2007, 2006, 2005)
2. (Klopfenstein and Erickson, 2002; Todd et al., 2011)
3. (Aarnink et al., 1995; Heber et al., 2000; Hoff et al., 2006; Jacobson and Hetchler, 2005)
4. (Arogo et al., 2003)
5. (Burns et al., 2007; Lacey et al., 2003)
6. (Casey et al., 2003; Coufal and Chavez, 2006)
7. (Fabbri et al., 2007; Nicholson et al., 2004)
8. (Liang et al., 2005; Nahm, 2003)
9. (Harper and Sharpe, 1998; Lim et al., 2003; Osada et al., 2000; Portejoie et al., 2003; Visscher et al., 2002; Zahn et al., 2000)
10. (Arogo et al., 2003)
11. (James, 2008; McGinn and Sommer, 2007)
12. (Chantigny et al., 2007; Sharpe and Harper, 1997; Westerman et al., 1995)
13. (Pelletier, 2008; Redwine et al., 2002)
14. (Sommer and Hutchings, 2001)

Table S4.1. State by state distribution of cattle on feed.

STATE	CATTLE ON FEED	CATTLE EXCL COWS	BEEF COWS	TOTAL BEEF	% ON FEED
ALABAMA	0	504,564	722,787	1,227,351	0.0%
ARIZONA	272,175	519,812	197,901	989,888	27.5%
ARKANSAS	235	793,552	813,250	1,607,037	0.0%
CALIFORNIA	488,131	2,971,282	583,594	4,043,007	12.1%
COLORADO	1,009,873	1,816,055	683,291	3,509,219	28.8%
CONNECTICUT	104	22,458	8,080	30,642	0.3%
DELAWARE	2,545	9,880	3,833	16,258	15.7%
FLORIDA	2403	569,313	982,790	1,554,506	0.2%
GEORGIA	0	484,283	469,942	954,225	0.0%
IDAHO	263,466	1,333,755	485,025	2,082,246	12.7%
ILLINOIS	276,130	684,809	343,972	1,304,911	21.2%
INDIANA	76,134	464,497	182,627	723,258	10.5%
IOWA	1,550,523	2,803,358	885,568	5,239,449	29.6%
KANSAS	2,255,701	4,519,961	1,270,538	8,046,200	28.0%
KENTUCKY	21,346	1,214,013	985,075	2,220,434	1.0%
LOUISIANA	0	338,626	434,252	772,878	0.0%
MAINE	2,631	43,634	10,505	56,770	4.6%
MARYLAND	7,851	104,413	39,188	151,452	5.2%
MASSACHUSETTS	442	16,963	6,240	23,645	1.9%
MICHIGAN	148,608	646,096	108,126	902,830	16.5%
MINNESOTA	536,971	1,591,546	357,826	2,486,343	21.6%
MISSISSIPPI	0	411,647	495,381	907,028	0.0%
MISSOURI	85,060	1,926,437	1,683,731	3,695,228	2.3%
MONTANA	52,345	1,180,140	1,439,653	2,672,138	2.0%
NEBRASKA	2,647,855	4,600,935	1,730,112	8,978,902	29.5%
NEVADA	2403	170,688	220,150	393,241	0.6%
NEW HAMPSHIRE	2403	15,843	4,075	22,321	10.8%
NEW JERSEY	362	14,757	9,500	24,619	1.5%
NEW MEXICO	44,936	573,767	461,595	1,080,298	4.2%
NEW YORK	26,976	722,623	86,030	835,629	3.2%
NORTH CAROLINA	2,137	435,561	348,196	785,894	0.3%
NORTH DAKOTA	58,408	910,055	881,682	1,850,145	3.2%
OHIO	164,487	696,487	277,949	1,138,923	14.4%
OKLAHOMA	353,923	2,522,182	1,677,903	4,554,008	7.8%
OREGON	84,657	667,899	504,279	1,256,835	6.7%
PENNSYLVANIA	128,732	945,790	148,249	1,222,771	10.5%
RHODE ISLAND	2403	2,011	1,447	5,861	41.0%
SOUTH CAROLINA	0	114,544	166,745	281,289	0.0%
SOUTH DAKOTA	418,374	2,190,861	1,610,559	4,219,794	9.9%
TENNESSEE	3,042	933,708	874,630	1,811,380	0.2%
TEXAS	2,750,818	6,395,478	4,329,341	13,475,637	20.4%
UTAH	23,857	316,714	369,670	710,241	3.4%
VERMONT	1,593	128,622	11,487	141,702	1.1%
VIRGINIA	20,010	880,457	657,320	1,557,787	1.3%
WASHINGTON	246,170	683,951	211,852	1,141,973	21.6%
WEST VIRGINIA	2,794	213,415	191,398	407,607	0.7%
WISCONSIN	270,342	1,975,688	248,305	2,494,335	10.8%
WYOMING	76,833	637,283	664,254	1,378,370	5.6%
NATIONAL TOTAL	14386189	51,720,413	28,879,903	94,986,505	15.1%

Table S4.2. State Animal Populations

State	Swine	Beef	Dairy	Layers	Broilers
AL	142555	1227351	9116	9435605	172955409
AZ	134000	717713	193621	44123	8451
AR	109316	1606802	8972	12545952	170380435
CA	111893	3554876	1815655	19000779	42268482
CO	727301	2499346	130736	4195691	19571
CT	4737	30538	17740	30912	79605
DE	5891	13713	4512	3133	43206514
FL	14915	1552103	123220	9386611	11031656
GA	153753	954225	79492	17445067	243463943
ID	14894	1818780	578761	655346	9639
IL	4630796	1028781	98849	4327311	115927
IN	3747352	647124	174141	25587222	6238623
IA	20455666	3688926	204757	52218870	1948950
KS	1886197	5790499	131688	91731	17851
KY	313360	2199088	71783	4308549	51189742
LA	6806	772878	16089	1910683	25061453
ME	8923	54139	32117	3531186	47252
MD	19869	143601	50923	2364942	64192426
MA	11151	23203	12500	153925	18137
MI	1099478	754222	376255	12676021	1125601
MN	7606785	1949372	463312	9693648	7765172
MS	401898	907028	14480	5593802	134479892
MO	2774597	3610168	92952	8276409	46880714
MT	173953	2619793	13947	464802	89862
NE	2992576	6331047	54628	9351688	908965
NV	2000	390838	29484	21209	3813
NH	3287	19918	13474	221446	28924
NJ	7901	24257	7192	1543699	19945
NM	1294	1035362	318878	66653	3928
NY	74671	808653	610712	5208831	591576
NC	8901434	783757	45960	13091384	148251469
ND	133653	1791737	17876	92754	24708
OH	2058503	974436	267857	28312692	12194024
OK	2304740	4200085	45885	3121799	38429952
OR	12693	1172178	125767	2420907	3294786
PA	1134957	1094039	532335	25147630	29248115
RI	1830	3458	1209	69662	13402
SC	224076	281289	15997	4231250	44296198
SD	1191162	3801420	91831	2450780	57635
TN	147795	1808338	47978	1675399	30400743
TX	800893	10724819	434928	20902244	107351698
UT	731666	686384	90449	3814859	5629
VT	3874	140109	134142	212397	48545
VA	239899	1537777	94105	2897238	38386310
WA	19861	895803	266989	7236128	7511065
WV	5873	404813	10095	1113238	14781332
WI	311651	2223993	1270091	5413563	7818682
WY	85432	1301537	6194	26612	4857
USA	65947807	80600316	9249674	3.43E+08	1506271608

SUPPLEMENTAL FIGURES

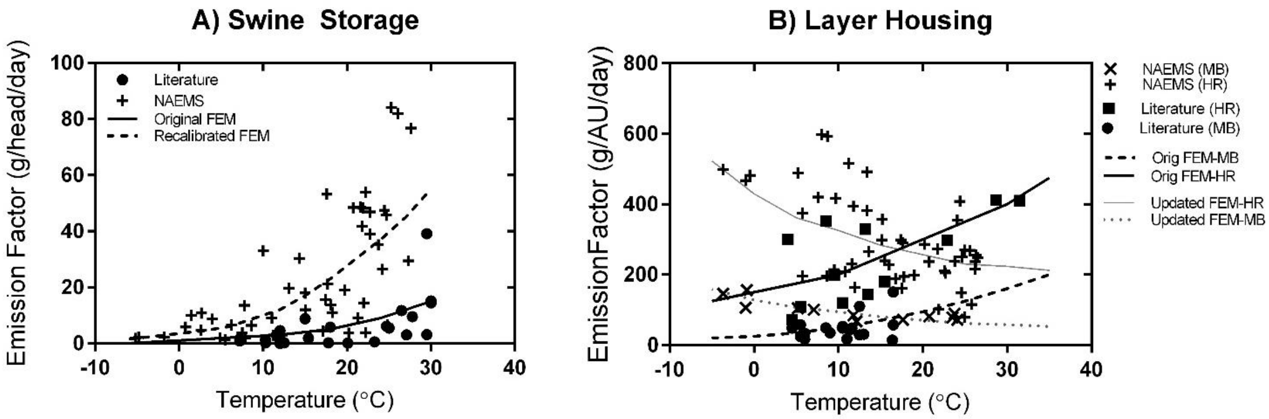

Figure S3.1: Model, literature and NAEMS Observations as a function of temperature with pre- and post-recalibration for NAEMS farms for a) swine storage and b) layer housing. Swine storage and layer housing had the most different emission factors reported from the literature and the NAEMS observations.

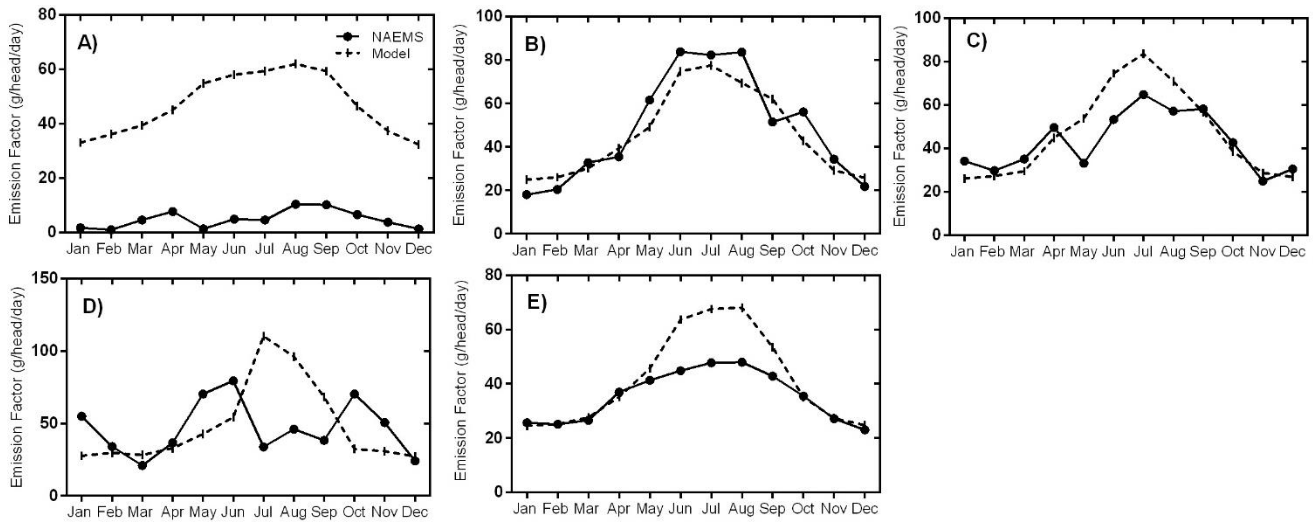

Figure S3.2: All measurement-model comparisons for seasonal model performance for dairy housing. Farms all have free-stall barns a) California, b) Indiana, c) New York, d) Washington, and e) Wisconsin.

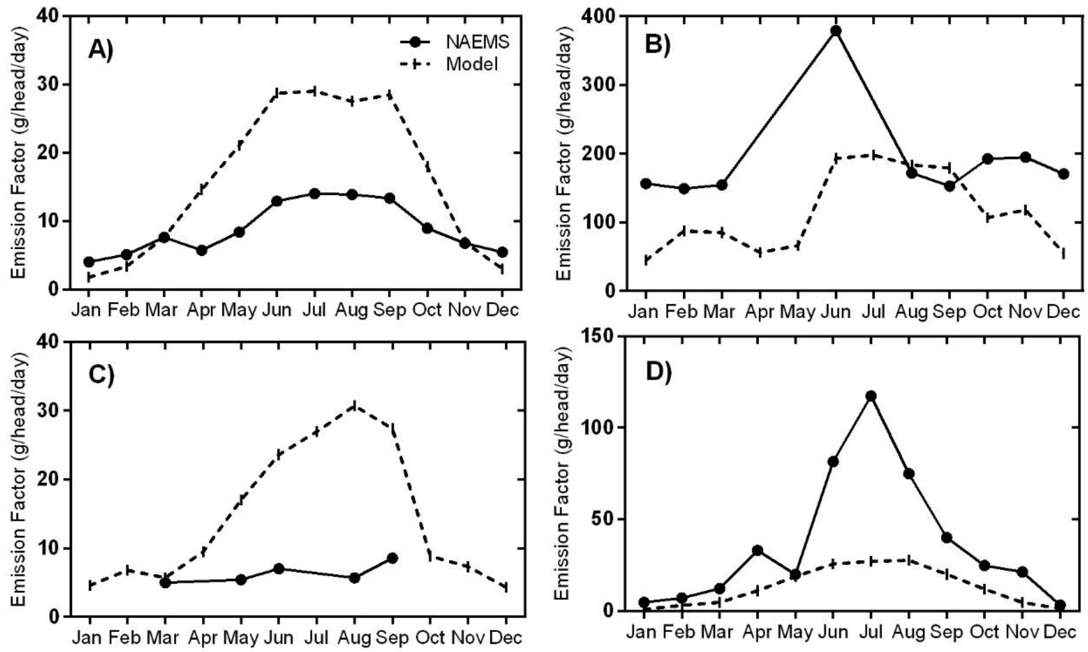

Figure S3.3: All measurement-model comparisons for seasonal model performance for dairy storage. Farms are of the following production stage and have the following types of housing a) IN5 (lagoon), b) TX5 (drylot), c) WA5 (lagoon), and d) WI5 (lagoon).

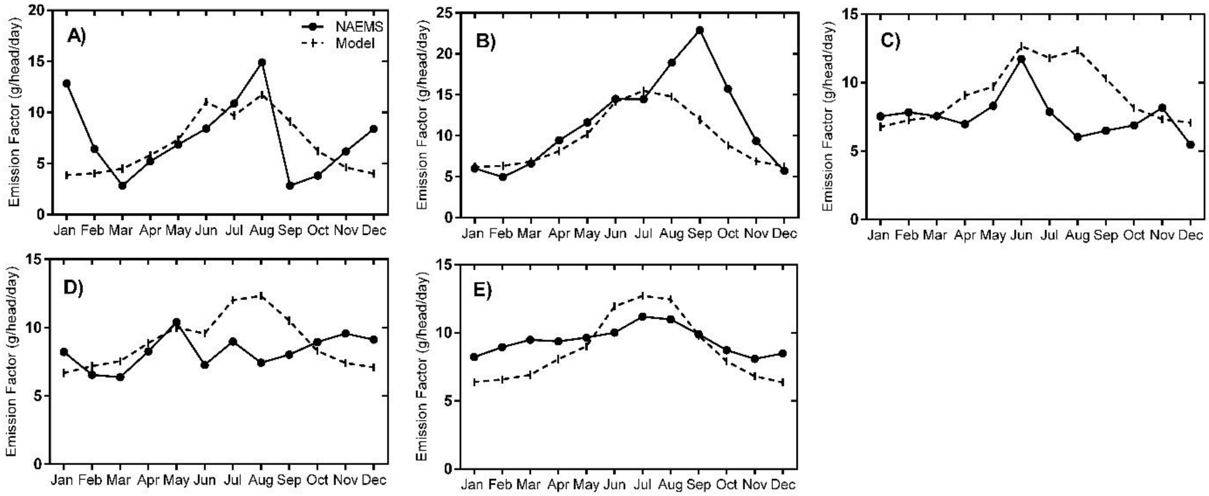

Figure S3.4: All measurement-model comparisons for seasonal model performance for swine housing. Farms are of the following production stage and have the following types of housing a) IN3 (deep-pit finishing), b) IA4 (deep-pit gestation), c) NC4 (flush gestation), d) NC3 (flush finishing, and e) OK4 (flush gestation)

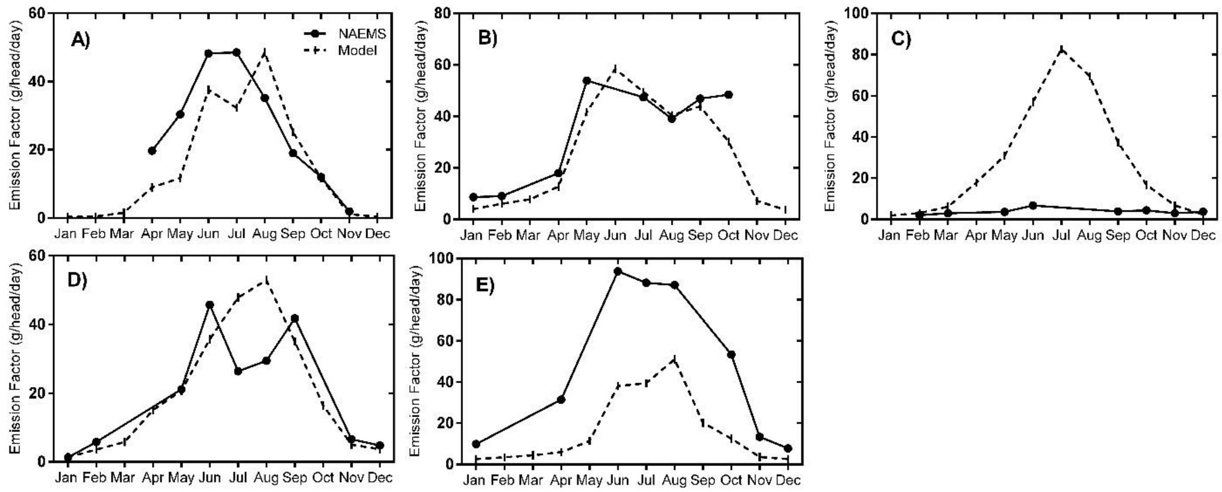

Figure S3.5: All measurement-model comparisons for seasonal model performance for swine storage. Farms are of the following production stage and have the following types of housing a) IN4 (lagoon), b) NC4 (lagoon), c) NC3 (lagoon), d) OK4 (lagoon), and e) OK3 (lagoon)

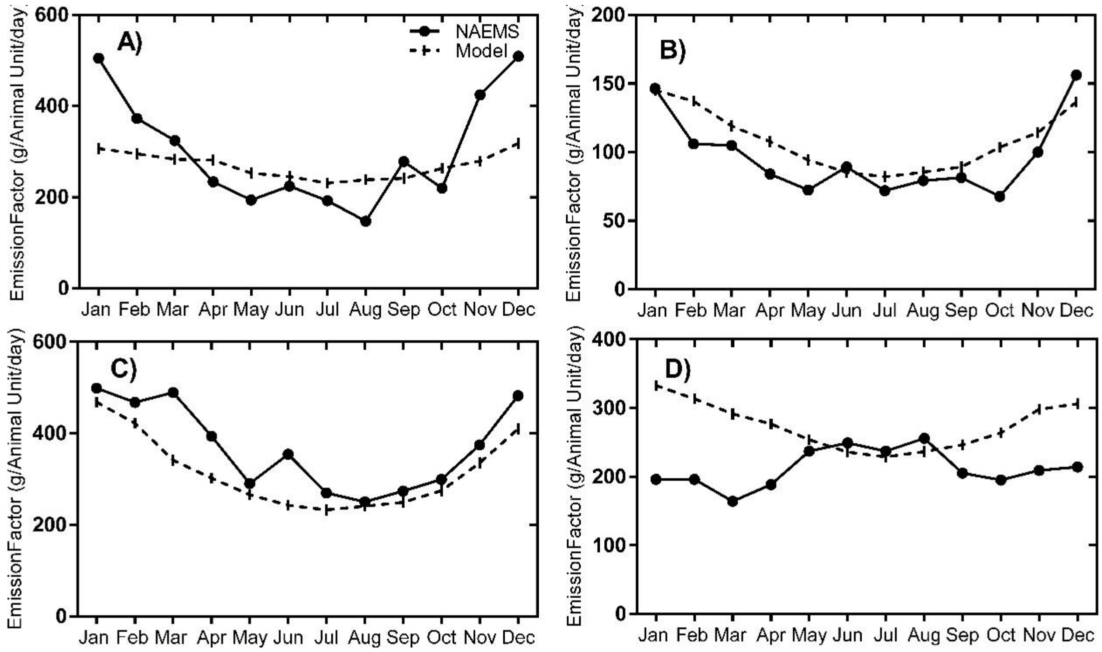

Figure S3.6: All measurement-model comparisons for seasonal model performance for layer housing. Farms have the following types of housing a) CA2 (high-rise), b) IN2B (manure-belt), c) IN2H (high-rise), and d) NC2 (high-rise).

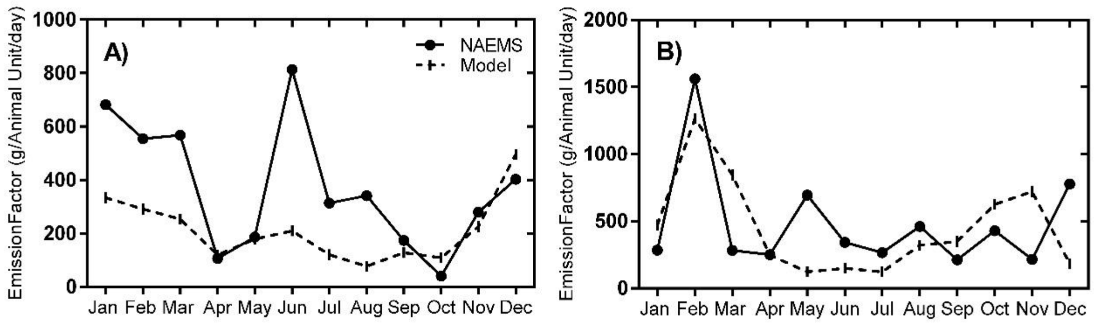

Figure S3.7 All measurement-model comparisons for seasonal model performance for broiler housing. Farms both have litter-based housing and are located in a) CA and b) KY.

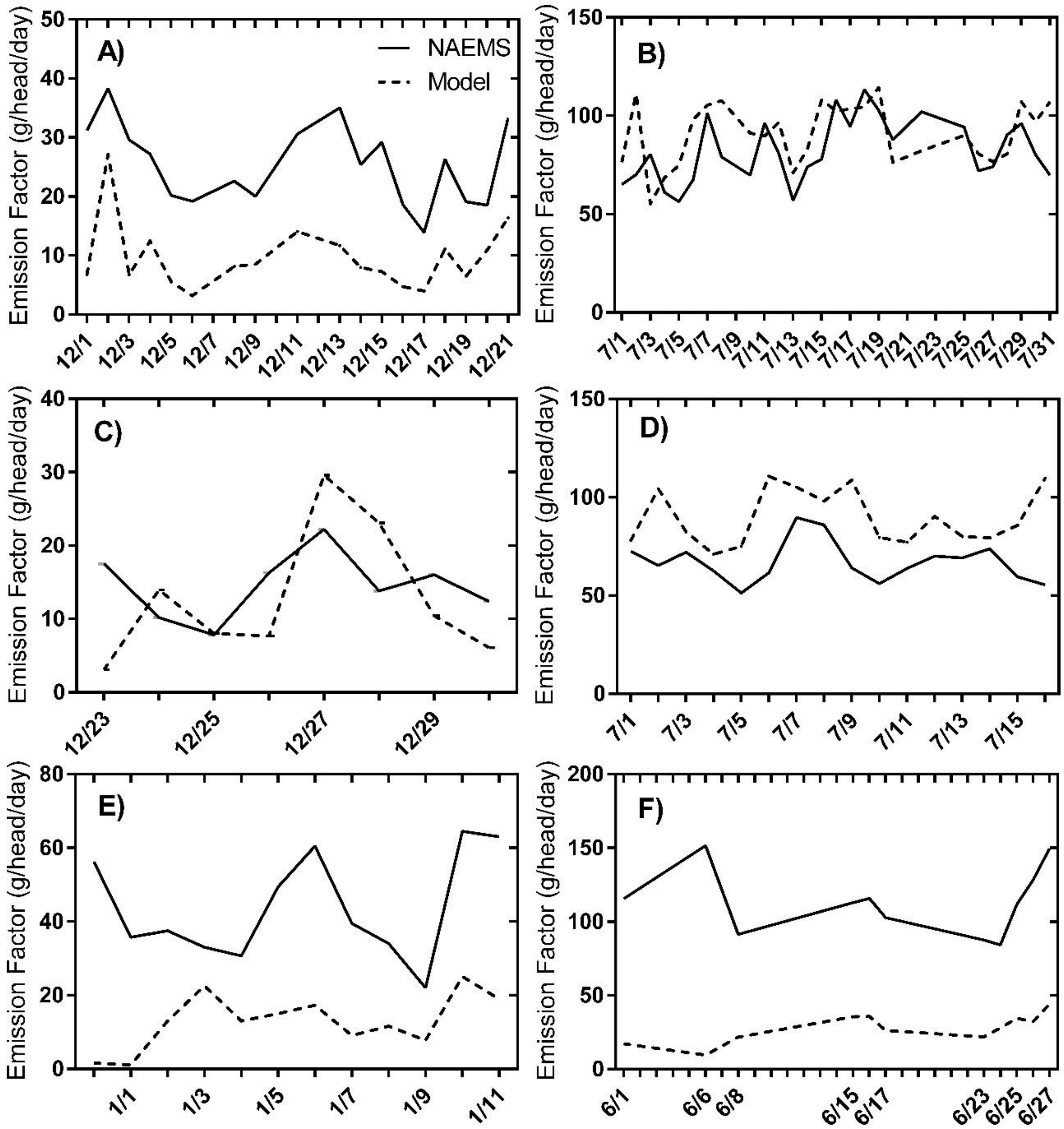

Figure S3.8: Daily variability in NAEMS data captured by model for: a free-stall dairy house in Indiana during a) December 2007 and b) July 2008, a free-stall dairy house in New York during c) December 2007 and d) July 2008, and a free-stall dairy house in Washington during e) January 2008 and f) June 2008.

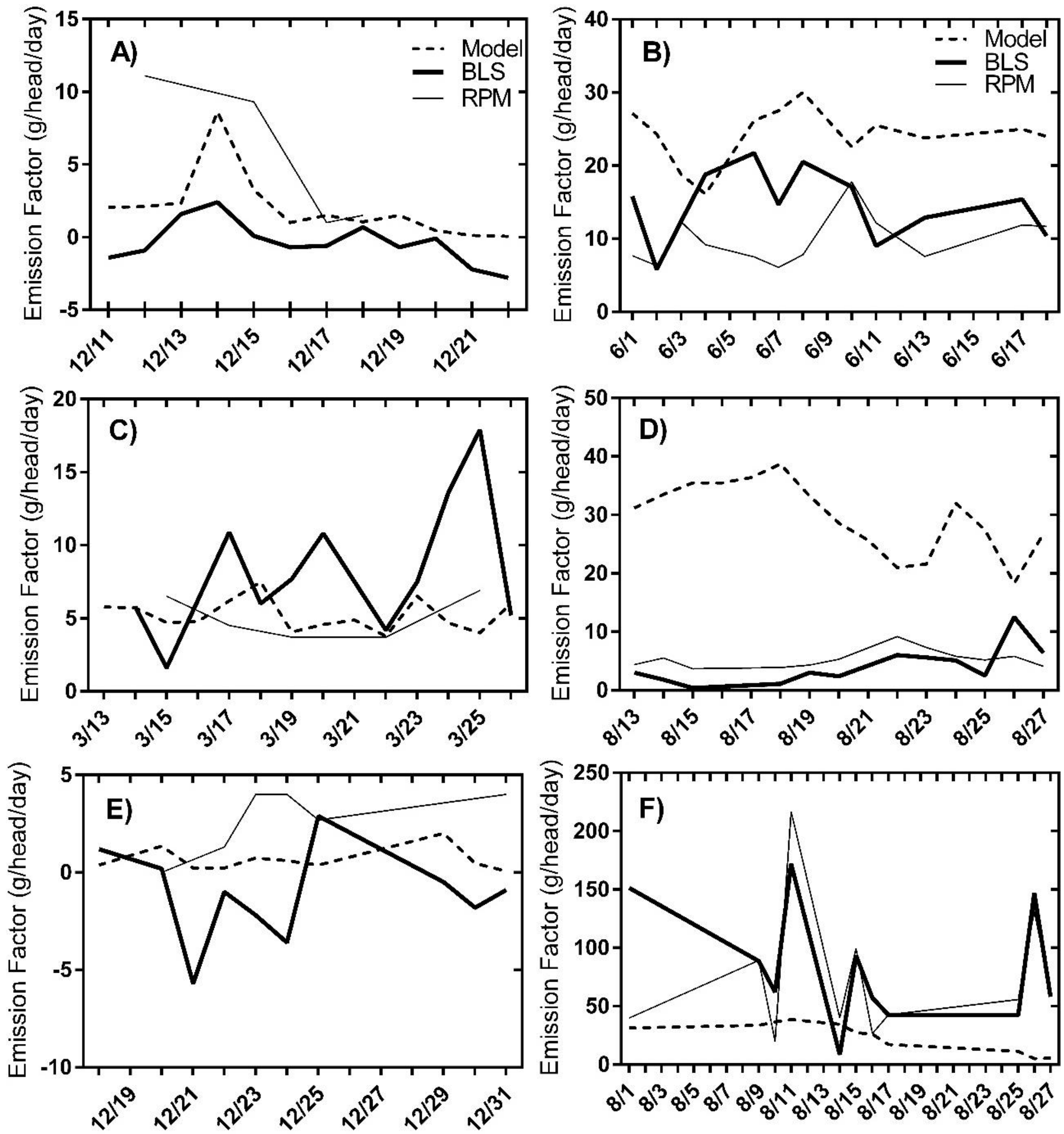

Figure S3.9: Daily variability in NAEMS data captured by model for: a dairy lagoon in Indiana during a) December 2008 and b) June 2008, a dairy basin in Washington during c) March 2008 and d) August 2008, and a dairy lagoon in Wisconsin during e) December 2008 and f) August 2008.

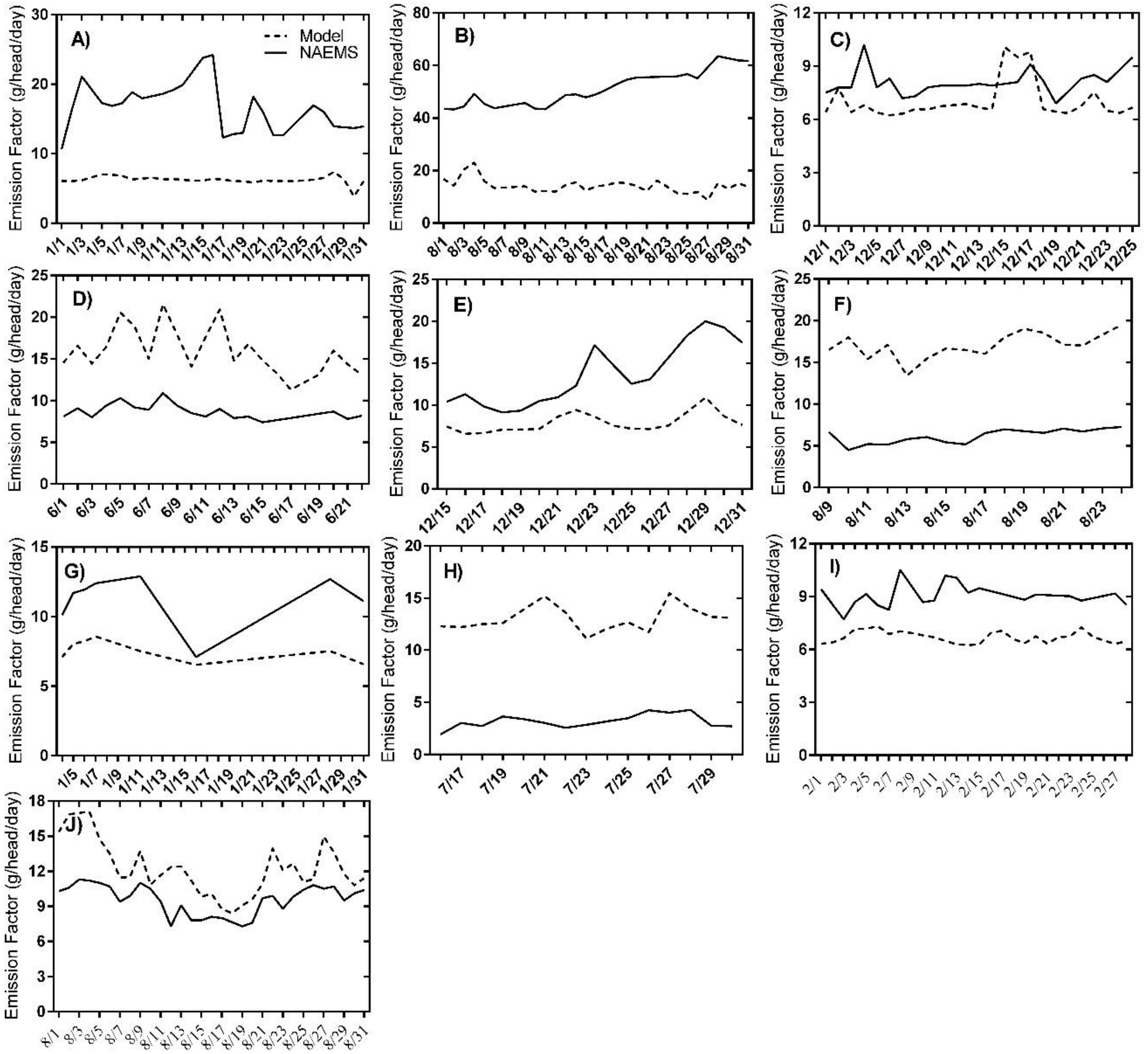

Figure S3.10: Daily variability in NAEMS data captured by model for: a deep-pit swine house in Iowa during a) January 2008 and b) August 2008, a deep-pit swine house in Indiana during c) December 2007 and d) June 2008, a shallow-pit swine house in North Carolina (NC3) during e) December 2007 and f) August 2008, a shallow-pit swine house in North Carolina (NC4) during g) January 2008 and h) July 2008, and a shallow-pit swine house in Oklahoma during i) February 2008 and j) August 2008.

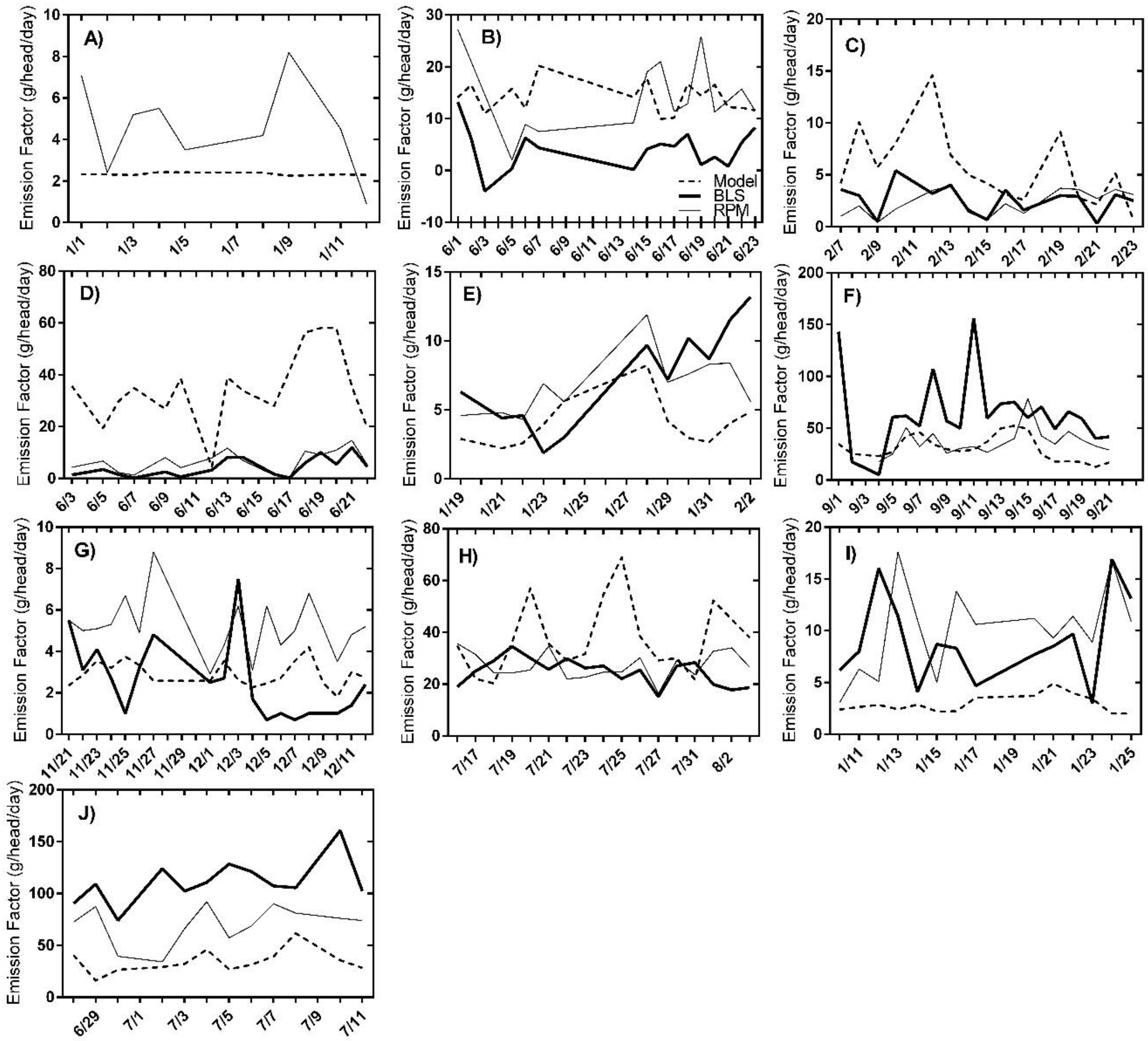

Figure S3.11: Daily variability in NAEMS data captured by model for: a swine basin in Iowa during a) January 2008 and b) June 2008, a swine lagoon North Carolina (NC3) during c) February 2008 and d) June 2008, a swine lagoon in North Carolina (NC4) during e) January 2008 and f) September 2008, a swine lagoon Oklahoma (OK3) during g) November-December 2008 and h) July 2008, and a swine lagoon in Oklahoma (OK4) during i) January 2008 and j) July 2008.

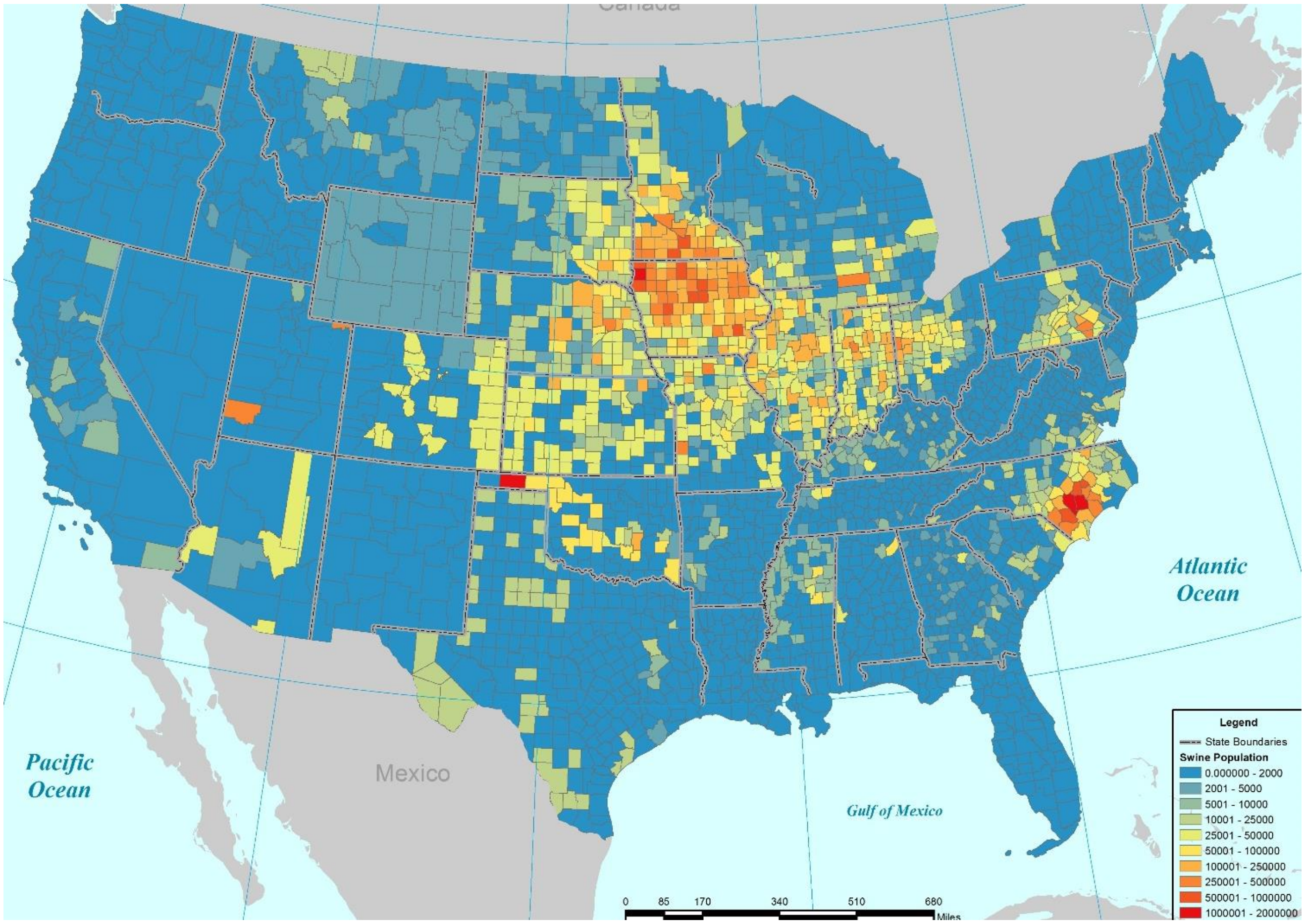

Figure S4.1: County-level swine population (2012)

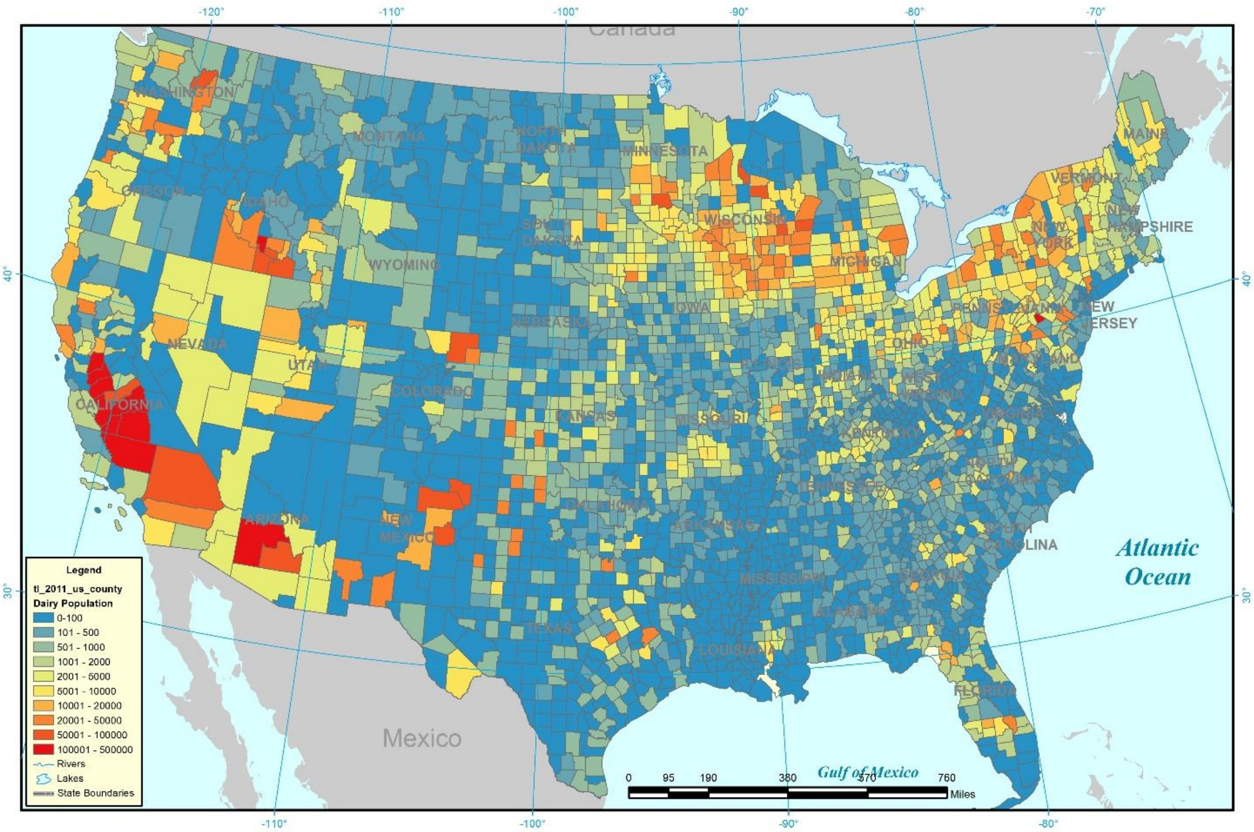

Figure S 4.2: County-level dairy cattle population (2012)

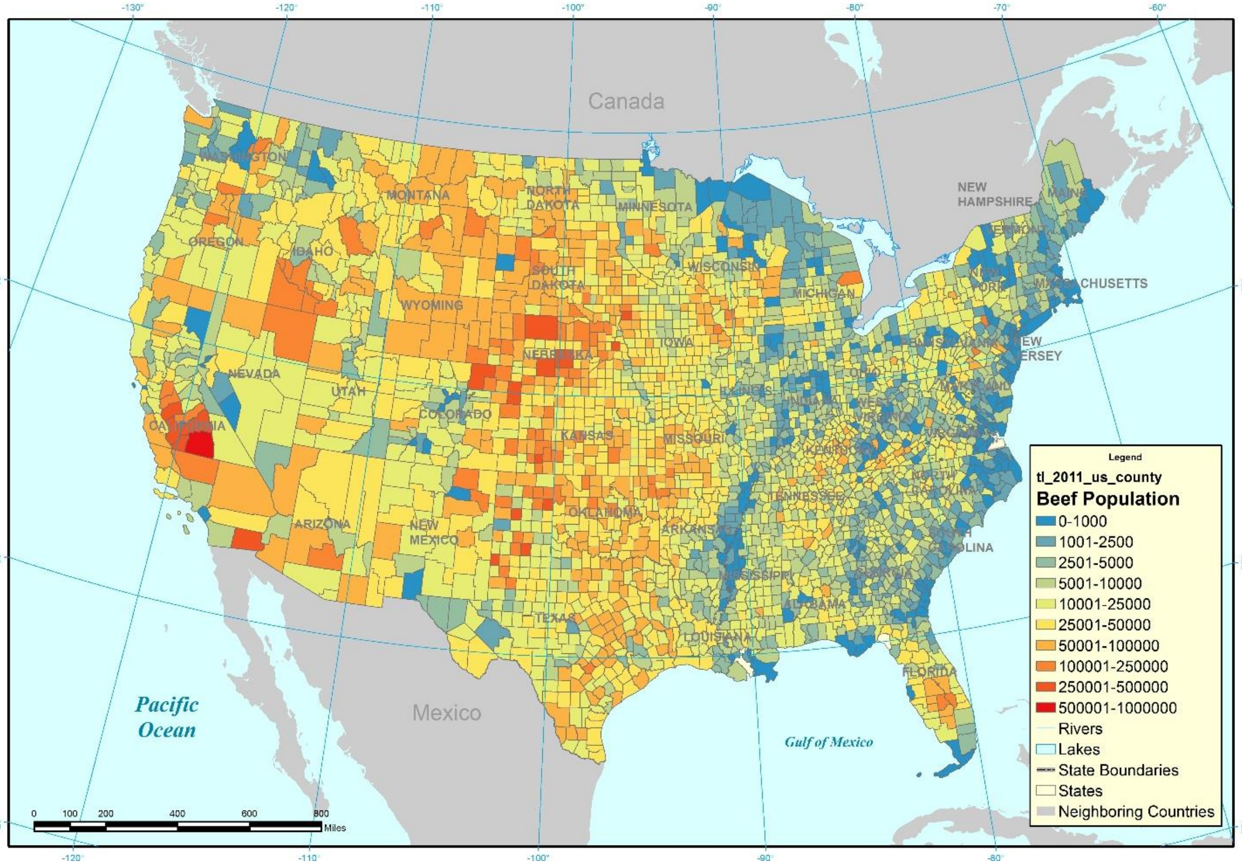

Figure S4.3: County-level beef cattle population (2012)

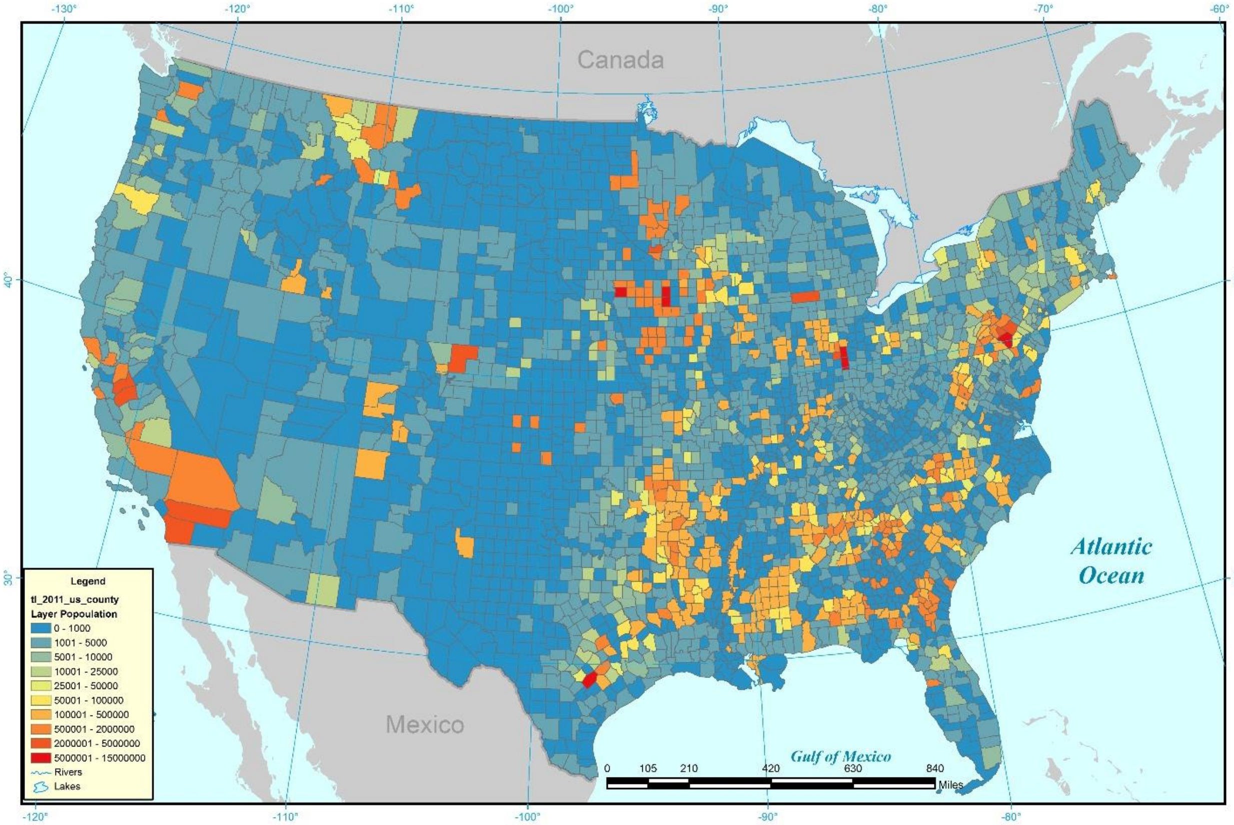

Figure S4.4: County-level layer chicken population (2012)

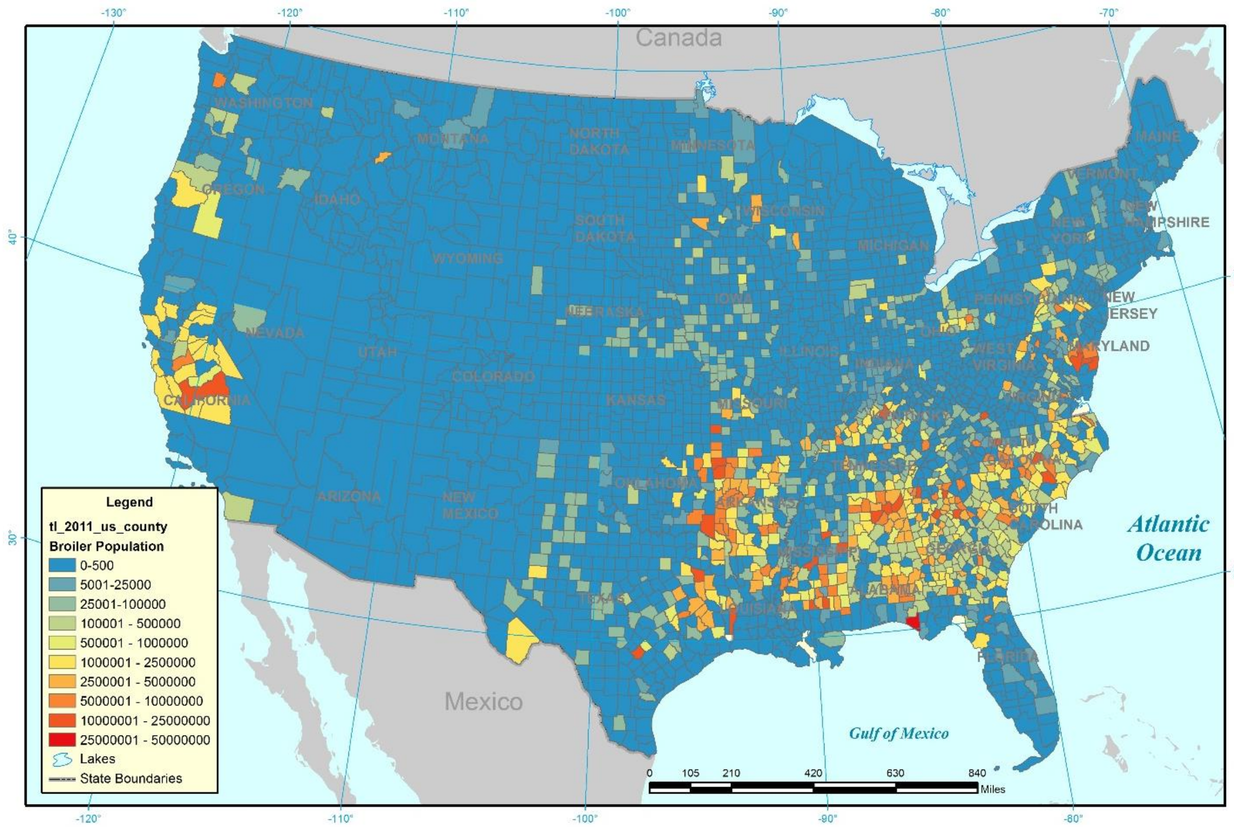

Figure S4.5: County-level broiler chicken population (2012)

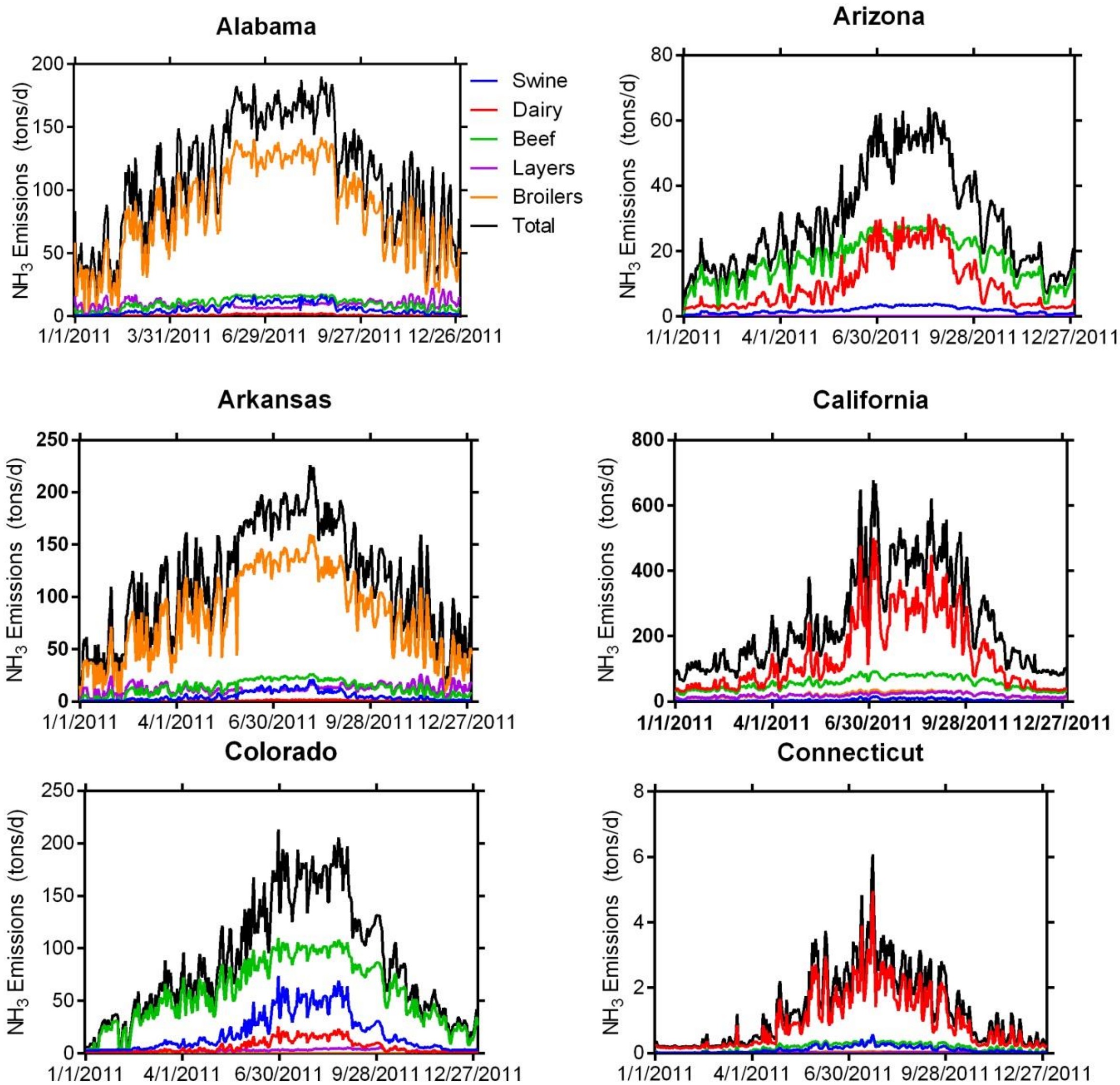

Figure S4.6: State livestock ammonia emissions (2011) by livestock type for Alabama, Arizona, Arkansas, California, Colorado, and Connecticut. Emissions are shown in tons NH3 per day.

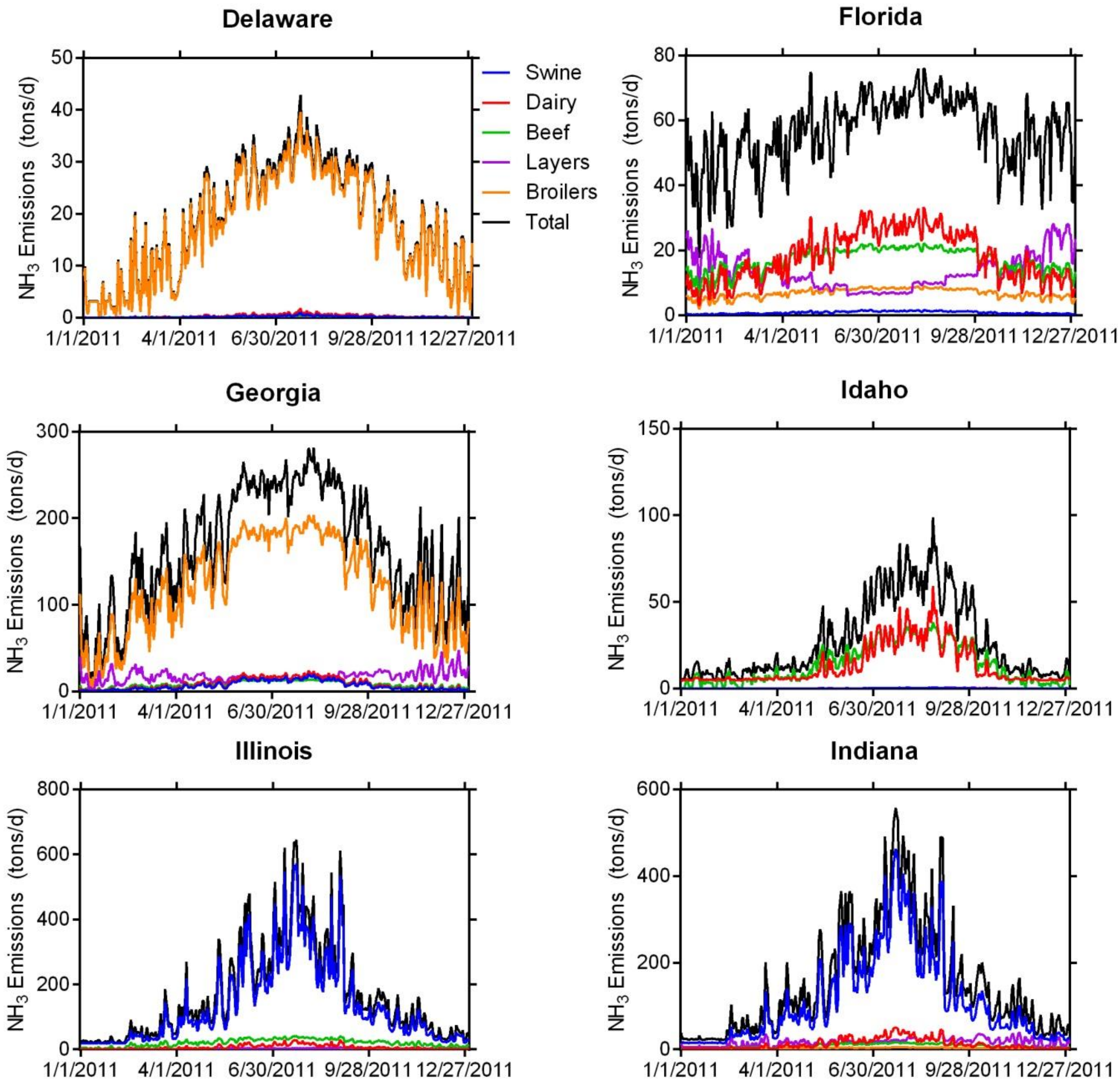

Figure S4.7: State livestock ammonia emissions (2011) by livestock type for Delaware, Florida, Georgia, Idaho, Illinois, and Indiana. Emissions are shown in tons NH_3 per day.

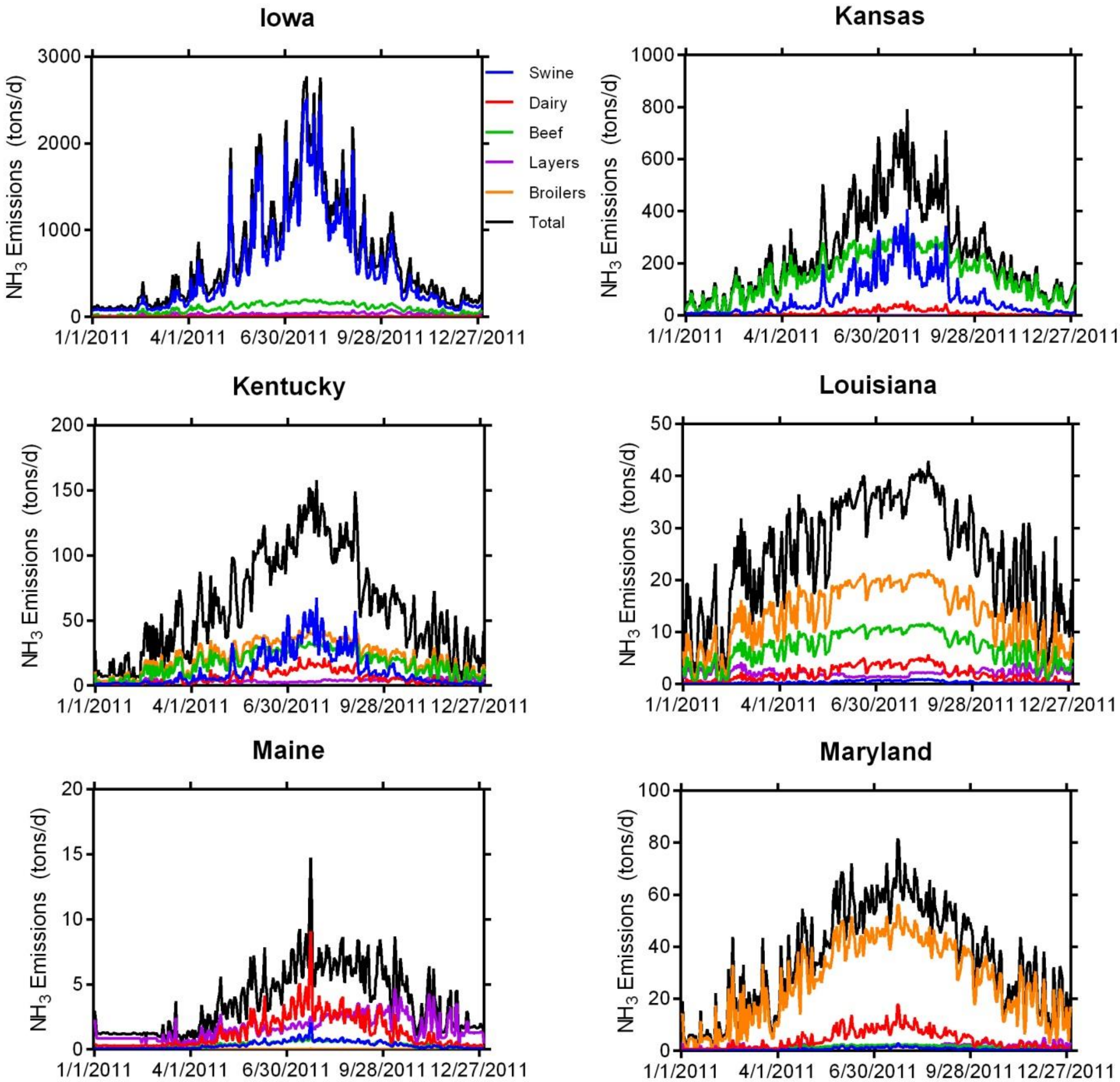

Figure S4.8: State livestock ammonia emissions (2011) by livestock type for Iowa, Kansas, Kentucky, Louisiana, Maine, and Maryland. Emissions are shown in tons NH_3 per day.

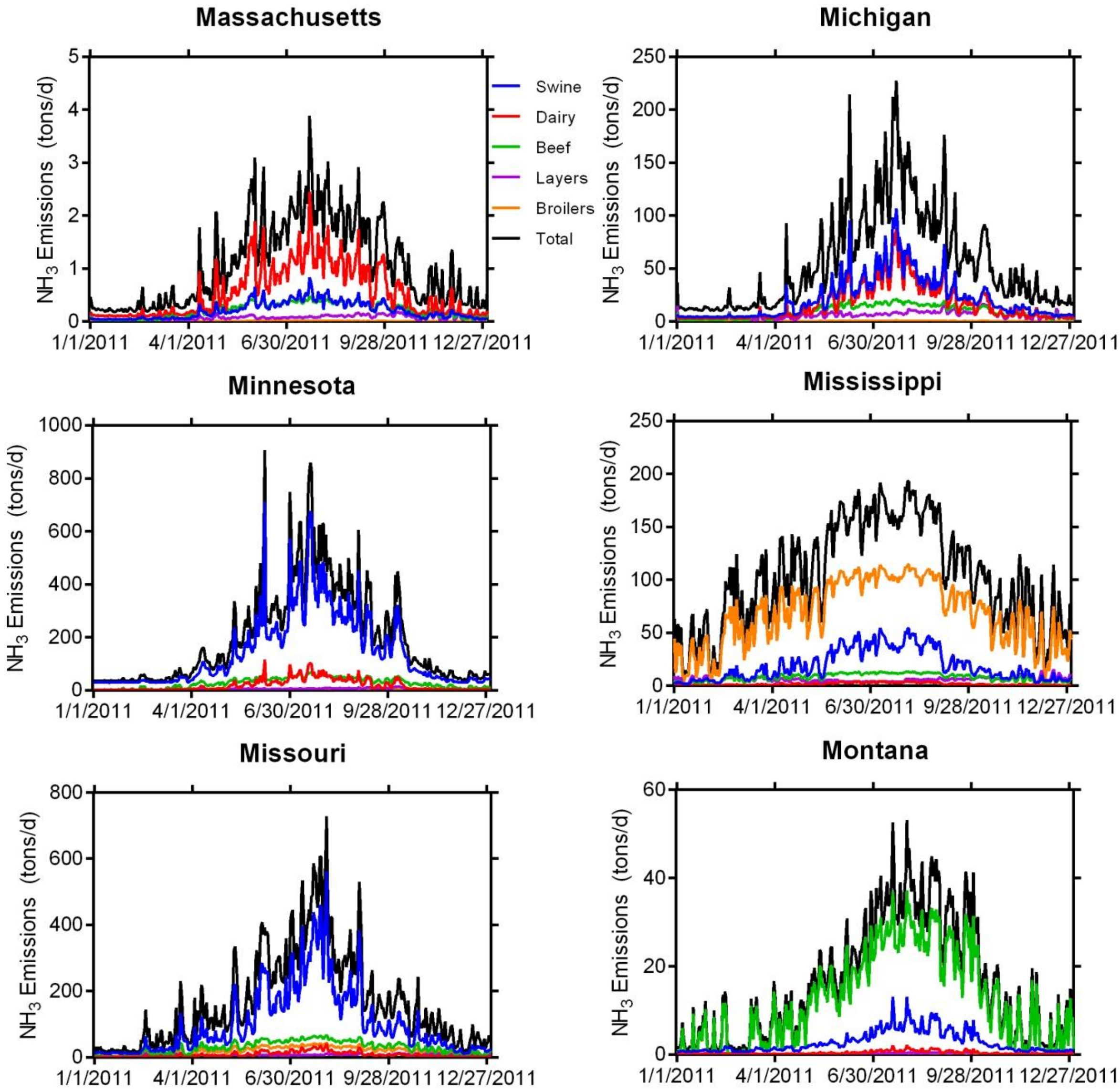

Figure S4.9: State livestock ammonia emissions (2011) by livestock type for Massachusetts, Michigan, Minnesota, Mississippi, Missouri, and Montana. Emissions are shown in tons NH_3 per day.

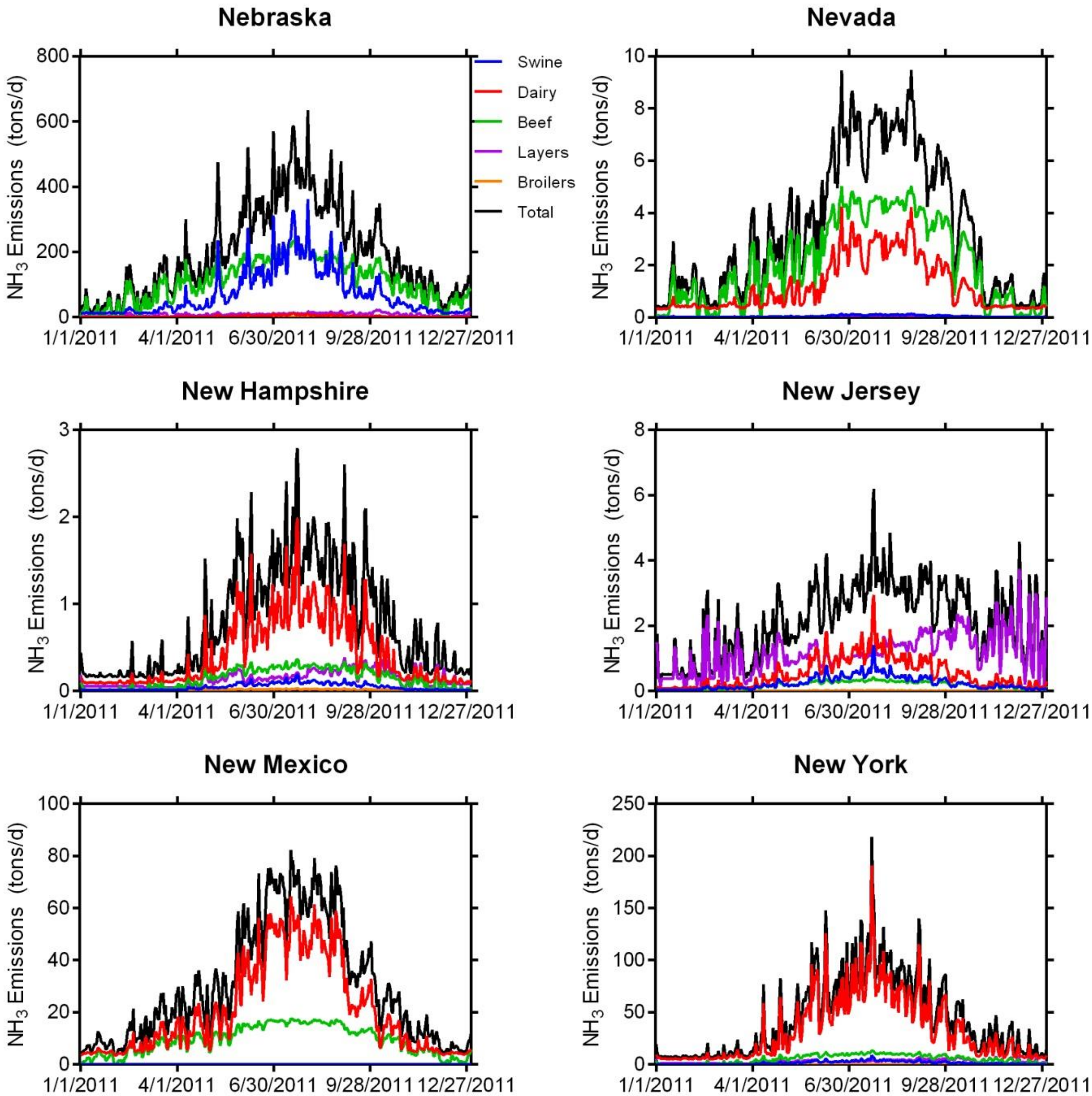

Figure S4.10: State livestock ammonia emissions (2011) by livestock type for Nebraska, Nevada, New Hampshire, New Jersey, New Mexico, and New York. Emissions are shown in tons NH_3 per day.

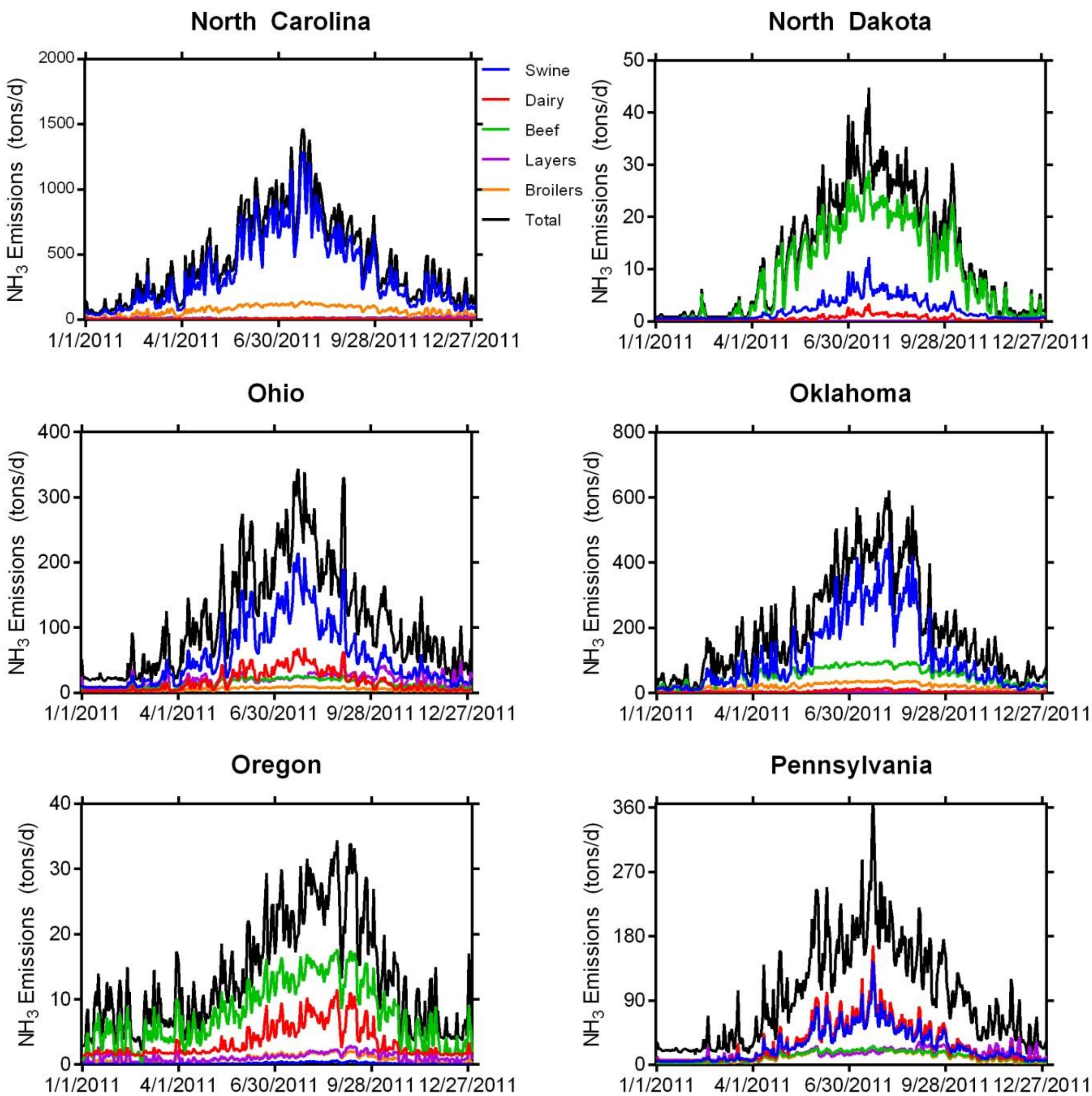

Figure S4.11: State livestock ammonia emissions (2011) by livestock type for North Carolina, North Dakota, Ohio, Oklahoma, Oregon, and Pennsylvania. Emissions are shown in tons NH_3 per day.

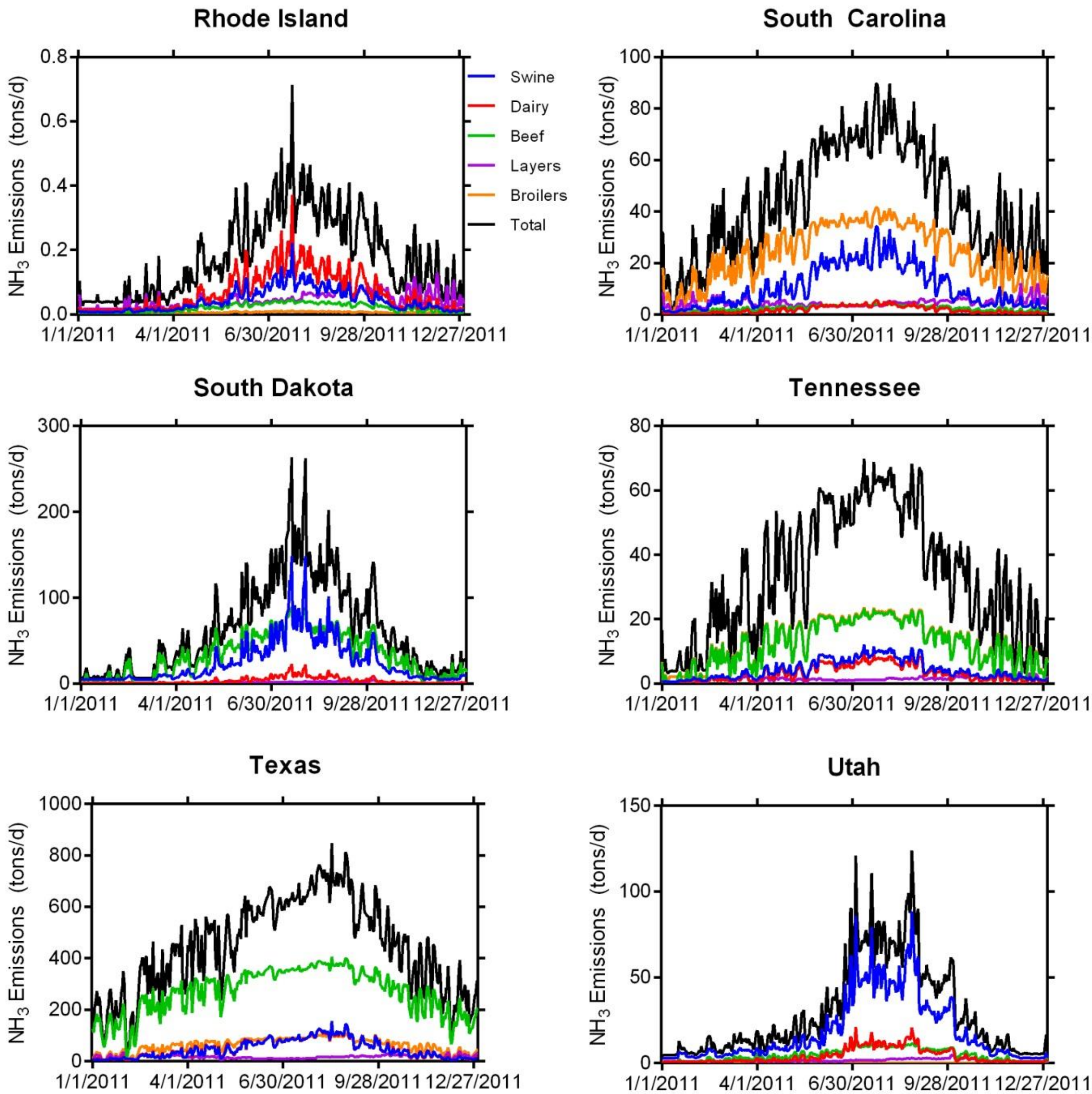

Figure S4.12: State livestock ammonia emissions (2011) by livestock type for Rhode Island, South Carolina, South Dakota, Tennessee, Texas, and Utah. Emissions are shown in tons NH_3 per day.

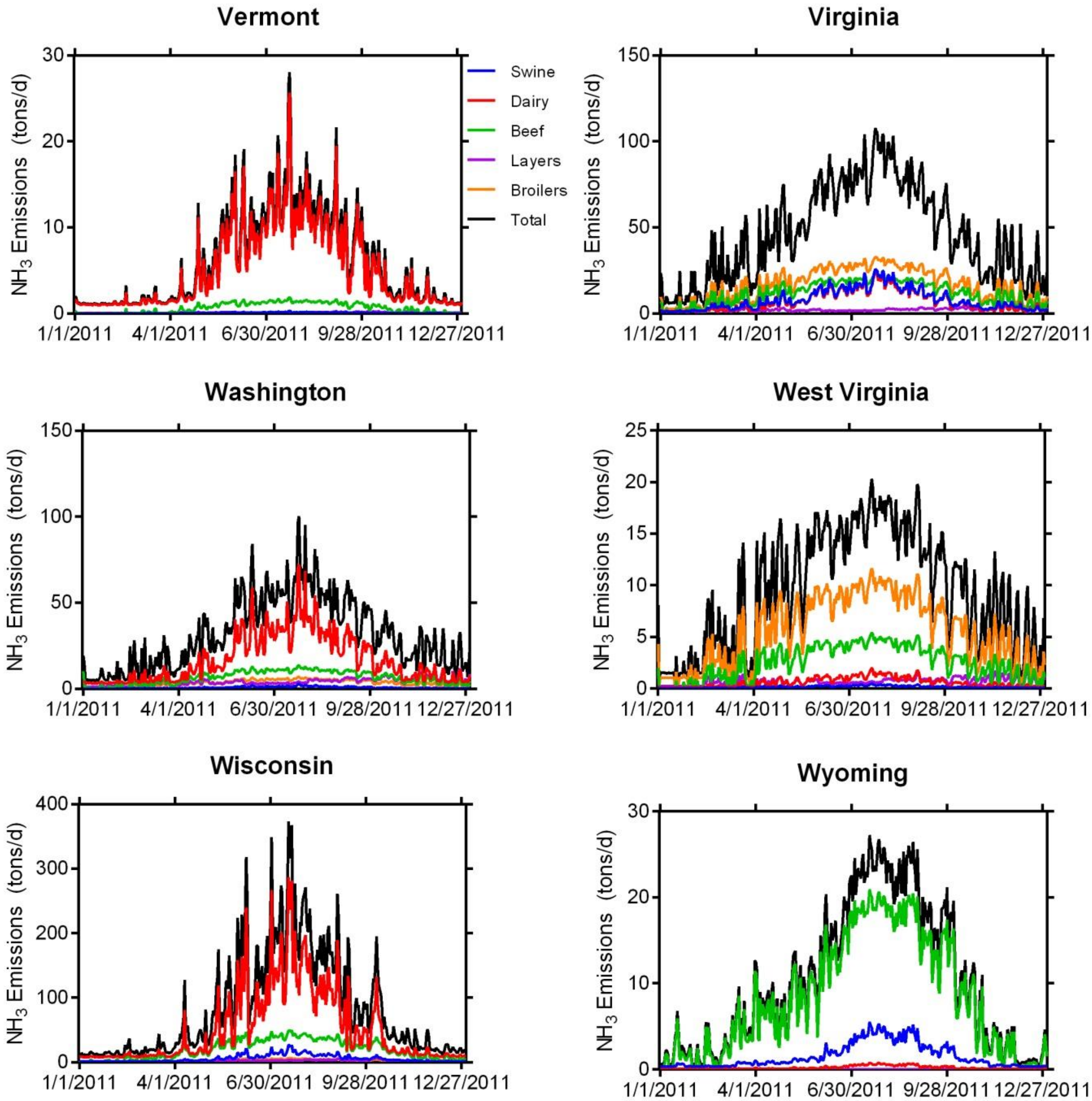

Figure S4.13: State livestock ammonia emissions (2011) by livestock type for Vermont, Virginia, Washington, West Virginia, Wisconsin, and Wyoming. Emissions are shown in tons NH_3 per day.