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A process-based model of ammonia emissions from dairy cows: improved temporal and spatial resolution

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Abstract

This research has developed an integrated model of a dairy farm that predicts monthly ammonia emission factors based on farming practices and climate conditions, including temperature, wind speed, and precipitation. The model can be used to predict the seasonal and geographic variations in ammonia emission factors, which are important for accurately predicting aerosol nitrate concentrations. The model tracks the volume of manure and mass of ammoniacal nitrogen as the manure moves through the housing, storage, application, and grazing stages of a dairy farm. Most of the processes of ammonia volatilization are modeled explicitly, but poorly understood processes are parameterized and tuned to match empirical data. The tuned model has been compared to independent experimental data and is shown to be robust over the range of experimental conditions. We have characterized the differences in emissions resulting from changes in climate conditions and farming practices and found that both of these factors are significant and should be included when developing a national inventory.

Keywords: Ammonia; Emission inventory; Agriculture; PM 2.5; Aerosol

1. Introduction

Ammonia is an important atmospheric pollutant that plays a key role in several air pollution problems. When combined with nitric acid, ammonia forms aerosol nitrate, which contributes significantly to total particulate matter (PM) (McNaughton and Vet, 1996). These particles have serious impacts on human health (Dockery et al., 1993) and influence climate (Charlson et al., 1992). When deposited in relatively pristine areas, an abundance of ammonia can cause degradation of aquatic (Jenkinson, 2001; Howarth et al., 2002) and terrestrial (Rennenberg and Gessler, 1999) ecosystems. While deposition of other atmospheric pollutants in the United States has been decreasing, concentrations of ammonia in precipitation have increased over the past 20 years (Nilles and Conley, 2001).

The sensitivity of ammonium nitrate aerosol concentrations to ammonia varies seasonally and geographically (West et al., 1999). However, previous inventories of ammonia emissions assume uniform emission factors for livestock across all locations and seasons (Pain et al., 1998; Hutchings et al., 2001). More accurate emission inventories require emission factors that are geographically and temporally resolved.

In both Europe and the United States, the largest source of ammonia emissions is livestock, estimated to account for 70–90% of total emissions, and dairy cows are one of the largest livestock sources (Battye, 1994; USEPA, 2000; Pain et al., 1998; Hutchings et al., 2001). These emissions arise from urine patches on grazed pastures, excreta deposited onto the floor of housing facilities, manure held in storage, and volatilization

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during the application of manure onto fields (Sommer and Hutchings, 1997).

Variation in ammonia emission factors results from the dependence of ammonia volatilization on meteorological conditions and seasonal and regional differences in farming practices. In field studies, high temperatures and wind speeds have been shown to increase the volatilization of ammonia (Sommer et al., 1991; Demmers et al., 1998). Heavy rains cause emissions to decrease to near zero (Sommer and Olesen, 2000). In cooler climates, cows are confined in housing units for the duration of winter; manure stored during this period is not applied to the fields until spring. In warmer climates, the cows may graze throughout the year. However, the amount of variation we can explain with current scientific understanding is limited. Because of the vast number of farm types and climate conditions, all of the experimental results to date cover only a small subset of possible emission scenarios.

We have addressed this need by developing the Farm Emissions Model (FEM), an integrated model of a dairy farm similar to that presented by Hutchings et al. (1996). The FEM predicts per cow emissions given a specific set of manure management practices and a temporal profile of temperature, precipitation, and wind speed. The FEM can be combined with the geographic distribution of manure management practices, animal populations, and climate data to produce an emission inventory. The results of such an application to the United States are discussed in a future paper.

To determine their model parameters, Hutchings et al. (1996) surveyed the literature and calculated the parameters based on the best available data. A major difference in our approach is that we use a formal technique to estimate model parameters. We apply Bayesian parameter estimation and experimental data to estimate the model parameters and uncertainty. The resulting parameters reflect the range of experimental conditions found in the literature. Where possible, we then validate our model with independent experimental results. As an example application of the FEM, we present monthly emission factors for four different farms in the United States.

2. Model description

2.1. Overview

Most experiments that measure ammonia emissions collect samples at a particular phase of the manure management process over a limited period of time. Such experiments generally focus on a subset of the factors that affect emissions. The FEM is designed to use these experimental results to generalize over the set of possible farming practices and conditions required for a large-scale emission inventory. In order to explain the variability present in emission factors, the FEM explicitly models the processes of ammonia volatilization that have the highest impact on emissions. However, some of the processes that govern volatilization are sufficiently uncertain that a mechanistic model cannot be justified, such as the effects of a surface crust on manure stored in an open tank or the effects of soil chemistry on the pH of field-applied manure. These processes are parameterized and tuned to match experimental data drawn from the literature. Factors that are known to be variable, but do not vary regionally or seasonally are assumed as constants.

The inputs to the FEM are a set of manure management practices and a temporal profile of climate conditions at a single farm, and the outputs are monthly average emission factors for that farm. The FEM tracks the flow of nitrogen through each of the stages of the manure management system: housing, storage, application, and grazing. Fig. 1 shows the flows of nitrogen in the model. Each stage has a separate submodel that accounts for the chemical and physical processes specific to that component. Mass of nitrogen and volume of manure are conserved throughout each submodel. In the first stage of the model, manure is partitioned between the housing and grazing submodels depending on the fraction of time the animal is housed. Manure deposited in housing structures is moved to storage daily. Solids may be separated from the manure and stored separately. Manure is moved from storage and applied to crops or pasture daily, weekly, monthly, or seasonally. While this framework could be extended to include emissions from calves and heifers, only mature dairy cows are considered here.

2.2. Nitrogen present in cow excreta

The nitrogen present in a cow's diet is partitioned between tissue growth, milk, urine, and feces. Urinary urea is the dominant source of volatilized ammonia and the only source considered by this model. On average, 75% of the nitrogen in urine is in the form of urea. The remainder of the urinary nitrogen is bound in other organic acids, some of which are thought to hydrolyze to

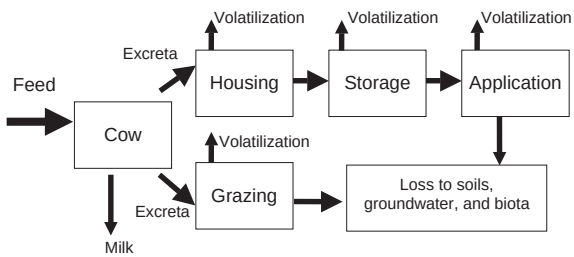

Fig. 1. Flows of nitrogen and volume of manure in the FEM.

ammonia (Bristow et al., 1992; Bussink and Oenema, 1998). These compounds are excluded because they are a minor fraction and their reaction pathways are not well understood.

Fecal nitrogen is separated in the model, because the majority of nitrogenous compounds found in feces are non-volatile. Fecal nitrogen is known to mineralize to form ammonia, but the contribution of this pathway is thought to be small (Haynes and Williams, 1993). On some smaller operations, urine, feces, and bedding are collected in the housing unit and stored in a heap or composted. Manure handled in this way maintains a solid consistency and is referred to as "solid manure". The seasonal variation in emissions from solid manure has not been well studied (Sommer and Hutchings, 2001), so a constant emission factor of $5\,\mathrm{kg\,cow^{-1}\,yr^{-1}}$ is assumed for emissions from storage and application, which is similar to published emission factors (Sommer and Dahl, 1999; Amon et al., 2001).

2.3. Transformations of nitrogen compounds

Urea nitrogen is hydrolyzed to form ammonium by urease enzyme, which is abundant in places inhabited by dairy cows (Jongebreur and Monteny, 2001). Ammonium then dissociates into aqueous phase ammonia and hydrogen ion, according to the dissociation equilibrium constant, K_a. Finally, ammonia is partitioned between the aqueous and gas phases according to Henry's Law constant, K_h. These are combined to yield the effective Henry's Law constant, H^*, (Eq. (1)) which determines the partitioning between gas phase ammonia and total ammoniacal nitrogen (TAN). Higher temperatures and higher pH favor gas phase ammonia.

$$H^* = \frac{[\mathrm{NH_3(g)}]}{[\mathrm{TAN}]} = \frac{K_a}{K_h[\mathrm{H^+}] + K_a(1 + K_h^{-1})}. \tag{1}$$

2.4. Processes affecting volume

The concentration of TAN is dependent on the solution volume, which is affected by precipitation, evaporation, and infiltration. Precipitation increases the volume of stored and recently applied manure while evaporation decreases the volume. Infiltration is the rate at which manure applied to fields or grazed pastures penetrates deep into the soil where it is no longer available for volatilization. This process simultaneously decreases both the volume of manure and the mass of TAN. Runoff of manure applied to fields and grazed pastures, while important for water quality models, is not considered here. Runoff is important during snowmelt and rain events (Kongoli and Bland, 2002; Walter et al., 2001), but in these conditions air emissions are low; therefore, it is unlikely that including this process will improve the model significantly.

2.5. Generalized description of ammonia volatilization

While there are structural and parametric differences between each of the submodels, they are similar in that ammonia is volatilized from the surface of a liquid solution and is then transported to the free atmosphere. The per cow volatilization of ammonia can be described as (Hutchings et al., 1996)

$$\mathrm{Emissions} = A[\mathrm{TAN}]H^* r^{-1}, \tag{2}$$

where A is the fouled surface area per cow ($\mathrm{m^2\,cow^{-1}}$), [TAN] is mass concentration of NH_3 and NH_4^+ in solution expressed as kg (as NH_3) $\mathrm{m^{-3}}$, H^* is the effective Henry's Law constant (dimensionless), and r is the mass transfer resistance ($\mathrm{day\,m^{-1}}$).

Mass transfer of gas phase ammonia is inhibited by the transport resistance, r. This resistance is the sum of the aerodynamic, quasi-laminar, and surface resistances. The aerodynamic resistance and quasi-laminar resistance result from resistance to transport in the turbulent layer above the slurry and the layer between the gas–liquid interface and the turbulent layer, respectively (Olesen and Sommer, 1994). Higher wind speeds cause these resistances to decrease. A complete description of the calculations for these resistances can be found in Olesen and Sommer (1994). The surface resistance arises from diffusion through the top layer of soil or a surface crust formed on the top of stored manure. It is meant to capture poorly understood processes that are specific to the stage of the model. This parameter is tuned to match empirical data.

Each submodel consists of three differential equations that govern changes in the manure volume, V ($\mathrm{m^3}$), [urea] (kg urea $\mathrm{m^{-3}}$), and [TAN].

$$\frac{\mathrm{d}V}{\mathrm{d}t} = k_{\mathrm{load}} + k_P A - k_E A - k_i A. \tag{3}$$

$$\frac{\mathrm{d[urea]}}{\mathrm{d}t} = k_{\mathrm{load}} C_{\mathrm{urea}} V^{-1} - k_{\mathrm{urea}}[\mathrm{urea}] - A[\mathrm{urea}]k_i V^{-1}. \tag{4}$$

$$\frac{\mathrm{d[TAN]}}{\mathrm{d}t} = k_{\mathrm{urea}} C_T[\mathrm{urea}] - A[\mathrm{TAN}]H^* r^{-1} - A[\mathrm{TAN}]k_i V^{-1}. \tag{5}$$

Manure volume is increased by the rate of manure loading, k_{load} ($\mathrm{m^3\,day^{-1}}$), and by precipitation: the per area rate of precipitation, k_P ($\mathrm{m\,h^{-1}}$), multiplied by the area (A). Evaporation and infiltration reduce the solution volume, with rate constants k_E and k_i ($\mathrm{m\,day^{-1}}$), respectively. Urea mass is added at the manure loading rate multiplied by the concentration of urea in newly deposited manure, C_{urea} (kg urea $\mathrm{m^{-3}}$). Urea is transformed to TAN by hydrolysis with rate constant k_{urea} ($\mathrm{day^{-1}}$), where C_T (kg TAN (kg urea)$^{-1}$) is the constant of stoichiometric conversion. TAN is

decreased by emissions and by infiltration. Transfers between submodels are assumed to occur instantaneously at prescribed intervals and are not shown in Eqs. (3)–(5).

2.5.1. Housing submodel

Because the manure is deposited indoors, the housing model differs from the other submodels in that the manure is not subject to precipitation, evaporation, or infiltration; the rate constants of these processes are equal to zero. Furthermore, previous studies have shown that ammonia volatilization in housing is related to the temperature of the ventilation air (Mannebeck and Oldenburg, 1991). The resistance parameter (in day m^{-1}) is not modeled as the sum of three separate resistance calculations; parameters H_1 and H_2 are tuned as a function of temperature.

$$r = H_1 + H_2 T. \tag{6}$$

Two types of housing structures are present in the model: freestall and tiestall. Cows in tiestall barns are confined to their stalls and are assumed to foul a smaller surface area.

2.5.2. Storage submodel

The storage model does not vary significantly from the generalized model described above. Manure is instantaneously transferred from the housing to storage once a day. For farms that apply manure monthly or more frequently, 90% manure is assumed to be transferred from storage during application. If the manure is applied seasonally, this fraction varies with each month. Manure is stored in relatively impermeable structures; therefore, no infiltration occurs. The precipitation surface area includes both the surface area of the storage unit and additional area to account for runoff. The surface resistance represents the potential formation of a viscous layer or crust on the surface of the slurry. A separate resistance parameter is tuned for open storage tanks with and without surface crust.

Three different types of storage are considered by this model: lagoon, slurry tank, and earthen basin. Each has a different surface area per cow, which is calculated from recommendations found in dairying manuals (MWPS, 1985).

2.5.3. Application submodel

Manure is transferred from storage to the application stage daily, weekly, monthly, or seasonally. Urea nitrogen is not considered by this submodel because all of the urea has been hydrolyzed in earlier stages. It differs from the previously discussed submodels in that the volume infiltrates into the soil with rate constant k_i. Also, a fraction of the manure is intercepted by the crop canopy. The ammonia in the intercepted manure is subject only to volatilization, so all of the ammonia is emitted to the atmosphere. The remaining fraction either volatilizes or infiltrates into the soil.

Three parameters are tuned in the application model: a surface resistance and a two-parameter function that approximates the effects of dry matter content on volatilization (Eq. (7)). The surface resistance represents the resistance to transport of gas phase ammonia through the top layers of the soil. Parameters A_1 and A_2 are used to approximate the interactions of the slurry and soil. This model considers only the effect of dry matter content (DMC) on the infiltration rate. Previous results have shown that the relationship between dry matter content and emissions is sigmoidal (Sommer and Olesen, 1991), so a functional form was selected that produces this behavior.

$$k_i = K_i \times 10^{(A_1 + A_2 \mathrm{DMC})}, \tag{7}$$

where K_i is the county-average soil permeability (m day^{-1}) on land used for agriculture, as reported by the MUIR soil survey database (1997).

Four different manure application techniques are included in the model: irrigation, broadcast, trailing hose, and injection. Irrigation application uses a spray gun or sprinkler system with narrow outlets and is modeled with a low dry matter content. A broadcast spreader applies manure with a wide spray nozzle or splash plate, and is modeled with a higher dry matter content. Trailing hose spreaders apply manure close to the soil surface, so the model assumes that a smaller fraction of the volume is intercepted by the crop canopy. Injection deposits the slurry beneath the surface; therefore the model assumes that a smaller fraction of the applied volume is susceptible to volatilization.

2.5.4. Grazing submodel

Manure deposited in the grazing model is subject to all of the processes described above in Eqs. (3)–(5). The grazing model has one tuned parameter to represent surface resistance. Grazing cows are on either drylots or pasture, each with its own tuned surface resistance.

2.6. Sources of input data

We calculated the concentration of nitrogen in the urine, the duration of grazing seasons, and the seasonal application schedule using survey data from the United States (NAHMS, 1996). We estimate the partitioning of consumed nitrogen between milk, urine, and feces using a statistical analysis by Castillo et al. (2000) using the national average milk production, which is 27.2 kg milk cow^{-1} day^{-1}. The daily output of urea nitrogen in the urine is estimated to be 256 g N cow^{-1} day^{-1} for a lactating cow. Assuming that 14.8% of the cows are non-lactating (NAHMS, 1996) and have urinary urea nitrogen production of 64 g N cow^{-1} day^{-1} (MWPS,

1985), the yearly nitrogen input to the model is 82.8 kg N $cow^{-1} yr^{-1}$.

The monthly variation of emissions in the model results from seasonal climate changes, seasonal confinement practices, and the timing of the manure application. Animals are confined in either the winter, the summer, both seasons, or neither. In the model, animals in farms that report grazing in the summer are assumed to be grazing on spring and fall days when the temperature is greater than 10°C, giving areas with warmer climates longer grazing seasons. Survey data (NAHMS, 1996) suggest that farms in counties with spring and fall average temperatures greater than 10°C are significantly more likely to have grazing cows. Non-confined animals are assumed to graze for 10 h a day.

Manure can be applied either daily, weekly, monthly, or seasonally. The fraction of the seasonally applied manure in each month is derived from survey data from the Pennsylvania Manure Storage Study (Thompson and Bowers, 1991). Seasonal manure applications primarily occur in the planting months of April and October.

2.7. Parameter tuning

The model parameters, shown in Table 1, are tuned to match empirical results; however, each ammonia emissions study in the literature is unique. While for some stages of the manure management system, the literature is in general agreement, in others experimental results are conflicting, suggesting a high level of uncertainty. A robust model should closely approximate experimental results over all conditions. Accordingly, the tuning procedure must incorporate the findings from the various experiments in a consistent way that preserves the level of experimental uncertainty.

This research uses Bayesian parameter estimation with Monte Carlo simulation (Sohn et al., 2000) to derive probability distributions for each tuned parameter using experimental data from the literature. This approach is superior to regression, because it captures the interaction of the inputs in a mechanistic manner. Briefly, this technique first assumes a probabilistic prior distribution for each parameter. Monte Carlo simulation is used to iteratively sample the prior distribution. For each experiment, the submodel inputs are assigned to the values reported in experiment design. If a range is reported for an input value, the range is sampled at every Monte Carlo iteration. If no value is reported for a required input parameter, a range from the literature is used. The submodels are used to predict the results of these experiments. The likelihood of the tuned parameter for each iteration is computed by calculating the level of agreement of the submodel result with the experimental data as in Sohn et al. (2000).

$$\text{Likelihood} = \frac{1}{\sqrt{2\pi\sigma_\varepsilon}} \exp\left(-\frac{1}{2}\left[\frac{O - Y_k}{\sigma_\varepsilon}\right]^2\right), \tag{8}$$

where O is the observed data, Y_k is the model prediction for the kth iteration, and σ_ε is the observational experimental error. Dividing each likelihood result by

Table 1
Emissions measurements used for tuning parameters in the FEM

Submodel	Tuned parameter	Description	Sources	Observations
Housing	H_1, H_2	Two parameter function relating ambient temperature to resistance	Elzing and Monteny (1997)	5
			Monteny and Erisman (1998)	6
Storage	r_c (no crust)	Surface resistance for storage with and without a surface crust	de Bode (1991)	5
	r_c (crust)		Sommer et al. (1993)	2
			Xue et al. (1999)	2
Application	A_1, A_2	Two parameter function relating soil permeability to infiltration rate	Sommer et al. (1991)	42
	r_c (application)	Surface resistance for manure applied to fields or pasture	Sommer and Christensen (1991)	15
			Menzi et al. (1998)	16
			Gordon et al. (2001)	8
Grazing	r_c (drylot)	Surface resistance for grazed pastures or drylots	Misselbrook et al. (1998)	6
	r_c (pasture)		Jarvis et al. (1989a)	7
			Jarvis et al. (1989b)	4

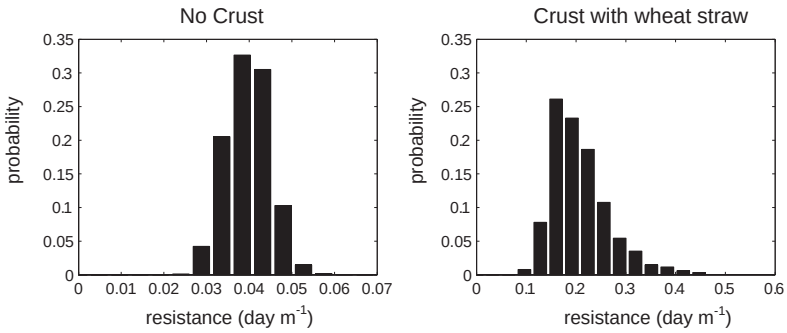

Fig. 2. Probability histogram for surface resistance for stored manure without a crust and with a crust amended with wheat straw. The crusted surface has a higher mean and greater variance, suggesting reduced emissions and greater uncertainty. The horizontal axis for the crust with wheat straw histogram is a factor of ten greater than the no crust histogram.

the sum of all the likelihood results yields the probabilistic posterior distribution of the parameter. The posterior distribution will have a high variance if the experimental data are sparse, noisy, or inconsistent. Hence, the uncertainty in the value of the tuned parameter is reflective of the ambiguities in the literature resulting from incomplete testing, difficulties in measurement, or contradictory results.

As examples of probability distributions of tuned parameters, Fig. 2 compares the storage submodel surface resistances for two cases: experiments that reported no crust formation on the surface of the stored manure and those that reported a wheat straw cover. When amended with wheat straw, stored liquid manure has a propensity to form a crust that can reduce emissions (Sommer et al., 1993). Accordingly, the mean resistance for experiments with a wheat straw cover is higher than the case with no surface crust. Also, the wheat straw case has greater variance, which can be attributed to more variable surface conditions.

When sufficient data were available, some datasets were reserved from the parameter estimation routines for testing. Fig. 3 contains plots comparing the model estimations and the reserved data for the storage, application and grazing submodels. Sufficient data were not available for the housing model, so the model is compared with one of the tuning sets. While the housing and storage models have reasonably small error, the application and grazing submodels have the largest error, possibly a result of insufficient detail in the modeling of soil interactions.

3. Results

Fig. 4 shows emissions from a confined and a grazing farm in two counties: Tulare County, California and Lancaster County, Pennsylvania. The confined California farm is typical of a large confined animal feeding operation, with manure stored in lagoons and applied seasonally. The grazing California farm has the same manure management system, but the animals graze seasonally. The Pennsylvania dairy with grazing is typical for a small farm with manure handled in solid form and applied daily, while the confined Pennsylvania dairy is identical to the confined California dairy. Temperature and precipitation data for these farms are derived from monthly distributions of daily averages compiled by the National Climate Data Center (NCDC, 2002a), and wind speed data (NCDC, 2002b) are derived from monthly averages.

The emissions exhibit a seasonal cycle, both from seasonal changes in farming practices and summer climate conditions that favor emissions. For the farm types in Fig. 4, the summer emissions are between 3 and 15 times higher than the winter emissions. Farms that have seasonal application also have high emissions in the planting months of April and October. Summer grazing decreases the summer emissions relative to housed livestock. While the farming practices have the largest effect on the emissions, the difference in climate is also evident. For the confined dairies, the warmer climate causes the California farm to have winter emissions twice as high as the Pennsylvania farm.

By implementing and tuning additional submodels, the FEM can estimate the emission reductions from ammonia control strategies. Two strategies for reducing emissions are requiring a wheat straw cover on stored manure and requiring an injection spreader for manure application. For the confined California farm described above, the wheat straw cover reduces total emissions by 11%, the injection application by 32%, and implementing both reduces emissions by 49%. The surface cover alone does not decrease emissions as much as when combined with injection because ammonia retained in the slurry is emitted during application. This result emphasizes the importance of using an integrated farm model for comparing control strategies.

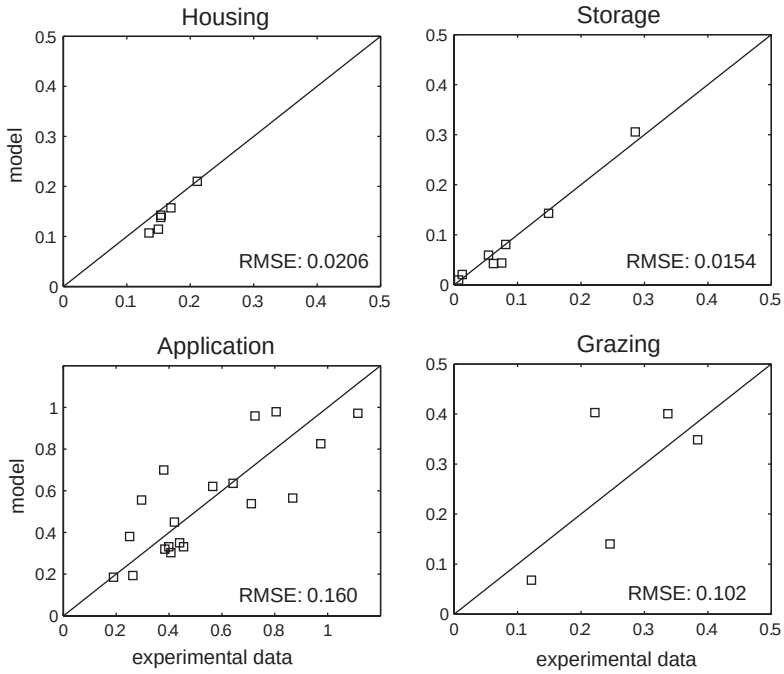

Fig. 3. Scatter-plot of fraction of input nitrogen volatilized as ammonia, showing a comparison of FEM predictions and experimental data for the following FEM submodels: housing, no-cover storage, application, and pasture grazing. Line denotes 1:1. The root mean squared error (RMSE) in units of fraction ammonia volatilized is shown in the lower right corner. The housing data is compared with the tuning set, while the other submodels are compared to independent experimental data. The storage phase is compared with Sommer (1997), the application phase is compared with Sommer and Olesen (1991), and the grazing phase is compared with Sherlock and Goh (1984).

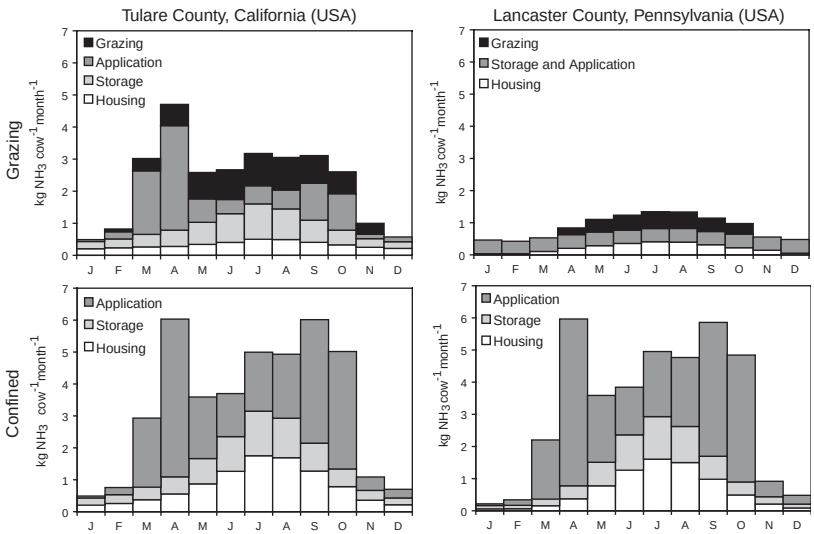

Fig. 4. Monthly average emission factors for different farming practices in California and Pennsylvania. The confined dairies have freestall housing where the animals are confined throughout the year, uncovered lagoon storage, and seasonal application of manure via irrigation. The grazing California farm is identical to the confined farm, except that the animals are not confined during the grazing season. The grazing Pennsylvania dairy has tiestall housing, manure is handled in the solid form, and the animals are not confined during the grazing season.

4. Conclusions

We have presented an integrated model for a dairy farm that predicts monthly ammonia emission factors for a variety of farming practices and climate conditions. Comparison with independent test data has shown good agreement. Our results suggest that farming practices are the most important determinant of annual average emissions. However, differences in the seasonal temperature, wind speed, and precipitation profile also significantly affect emissions. When calculating emission inventories, it is important to account for both farming practices and climate conditions.

Because the model presented here captures the dependence of ammonia emissions on manure management practices and climate conditions, it can be used to develop emissions inventories with high spatial and temporal resolution. This model has been combined with a database of farming practices and climate conditions to calculate a national inventory for the United States. These results and analysis of the sensitivity of the parameters of the FEM will be presented in a future paper.

Disclaimer

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