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Article

Improving Ammonia Emission Modeling and Inventories by Data Mining and Intelligent Interpretation of the National Air Emission Monitoring Study Database

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Abstract: Ammonia emission is one of the greatest environmental concerns in sustainable agriculture development. Several limitations and fundamental problems associated with the current agricultural ammonia emission modeling and emission inventories have been identified. They were associated with a significant disconnection between field monitoring data and knowledge about the data. Comprehensive field measurement datasets have not been fully exploited for scientific research and emission regulations. This situation can be considerably improved if the currently available data are better interpreted and the new knowledge is applied to update ammonia emission modeling techniques. The world's largest agricultural air quality monitoring database with more than 2.4 billion data points has recently been created by the United States' National Air Emission Monitoring Study. New approaches of data mining and intelligent interpretation of the database are planned to uncover new knowledge and to answer a series of questions that have been raised. The expected results of this new research idea include enhanced fundamental understanding of ammonia emissions from animal agriculture and improved accuracy and scope in regional and national ammonia emission inventories.

Keywords: air quality; animal agriculture; atmosphere; emission factor; pollution; process-based model

1. Introduction

Ammonia (NH_3) is a common substance playing an important role in the nitrogen cycle. In the boundless complexities of environmental interrelationships, ammonia is both a “friendly” molecule and a “hazardous” one [1]. Whether ammonia is friendly or hazardous depends largely on its concentrations in the atmosphere. Ammonia is the only base in the gas phase in the atmosphere, where it neutralizes acids at low concentrations. Ammonia emissions from agriculture can increase its concentration in the atmosphere to an excessive level. This has caused direct and indirect damage to the ecosystem in some regions with intensive animal production [2,3].

Moreover, ammonia from agriculture is a critical precursor of regional and national inhalable aerosols ($\text{PM}_{2.5}$) [4]. Anthropogenic sulfur emissions peaked in 1980 and have since begun to decrease [5]; therefore, ammonium nitrate will become a more important contributor to atmospheric $\text{PM}_{2.5}$ concentrations in some places. Reduction of ammonia emission may become a cost-effective strategy to reduce atmospheric $\text{PM}_{2.5}$ [6].

In modern animal agriculture, especially on poultry, cattle, and swine farms, animals are raised in concentrated animal feeding operations (AFO) and large quantities of manure are generated on the farms. Ammonia is produced from the manure during microbial processes that convert nitrogen in the manure into ammonia, which can be released from the manure and emitted from animal buildings to the outdoor atmosphere. High densities of animals in AFOs can result in high aerial ammonia concentrations in and large quantities of ammonia emissions from the animal buildings.

High concentrations of ammonia inside animal houses present potential health hazards to humans and animals [7,8]. There is still a great need to evaluate health effects of exposures to air pollutants including toxic gases emitted into the general environment from the AFOs [9].

In the past decades, emissions of ammonia have significantly increased as a result of intensive agricultural management and greater livestock production in Canada [10], Denmark, France, and the Netherlands [11], China [12], and many other developed and developing countries [13]. Therefore, agricultural ammonia emission has become one of the major worldwide air pollution concerns and has attracted increasing attention from the general public and government regulators. The European Parliament and the Council on National Emission Ceilings for certain pollutants (NEC Directive) set upper limits for each Member State for the total emissions in 2010 of four pollutants, which included ammonia [14]. Ammonia from agriculture production is one compound that would trigger the 45.4 kg per day reportable quantity of single “hazardous substance” by the Emergency Planning and Community Right-to-Know Act (EPCRA) in the U.S. [15,16].

Ammonia emission modeling and inventories provide emission estimates for types, amounts, locations, and timing of ammonia sources. The ultimate goal of the ammonia emission inventories is to identify emission patterns, plan control strategies, and achieve air quality standards. Present and future year inventories are used by scientists and decision makers to understand and improve air quality

through planning and modeling. The estimated ammonia emission from U.S. animal agricultural operations was 2,270,091 tons in 2002 using current ammonia emission inventories [17]. Yet the current ammonia emission inventories are highly uncertain [18] and emission models are insufficient for scaling up observations to larger areas and for testing mitigation strategies.

The objectives of this paper are to: (1) study the state-of-the-science of agricultural ammonia monitoring, modeling, and inventories, (2) identify research needs, (3) propose new research ideas and approaches, and (4) discuss expected results.

2. State-of-the-Science and Research Needs

Several limitations and fundamental problems associated with current agricultural ammonia emission modeling and inventories have been identified and need to be solved in a timely manner.

2.1. Data Integration

A significant disconnection between the field monitoring data and the ammonia emission factors and inventories has been identified. Agricultural air quality studies have experienced revolutionary changes in the past decades, especially after the introduction of advanced analytical instruments and personal computers [19]. Enormous amounts of measurement data have been collected in on-farm monitoring. So far there have been nine long-term (>6 months) and continuous field measurements monitoring at a total of 68 animal buildings in 12 states in the U.S. that have produced 3.3 billion comprehensive field sampling data points, each data point being a 1-min or 30-sec recorded value for one variable (Table 1).

Table 1. Reported long-term (>6 months) and continuous on-farm ammonia emission monitoring.

Year [a]	Scale and facility of study [b]	TDP [c]	IT [d]	Reference
1994–1995	4 MV finishing swine rooms for 6.5 mo, Belgium	1.1 M	12-min	[20]
1997–1998	4 NV and 4 MV finishing swine barns in 2 states for 6 mo, USA	155 M	20-sec	[21]
2001–2002	1 MV layer house for 6 mo, USA	18 M	1-min	[22]
2002–2003	2 MV pig finishing houses for 1 yr, USA	67 M	1-min	[23,24]
2003	10 MV layer houses in 2 states for 550 house-d, USA	26 K	30-min	[25]
2003–2004	12 MV pig and layer barns in 6 states for 1 yr, USA	200 M	1-min	[26]
2003–2004	3 pig compartments for 10 mo, Austria	NA	NA	[27]
2004–2008	3 MV layer houses, 1 house for 6 mo and 2 houses for 18-mo, USA	205 M	1-min	[28,29]
2006–2007	2 TMV broiler houses for 13 mo, USA	86 M	30-sec	[30]
2007–2008	4 MV layer barns and 1 manure shed in 2 states for 1 yr, USA	107 M	1-min	[31]
2007–2008	1 MV turkey barn for 1 yr, USA	83 M	30-sec	[32]
2007–2009	35 MV and NV broiler, layer, swine, and dairy buildings and 1 NV layer manure shed on 14 farms in 7 states for 2 yr, USA	2.4 B	1-min	[33]

Note: Adapted from Ni *et al.* [19]. [a] Years when monitoring was conducted. [b] MV=mechanically ventilated; NV=naturally ventilated; TMV=tunnel mechanically ventilated. [c] Total data points. NA=not available. [d] Integration time for each data point.

Despite the improvements in air quality monitoring, our ability to integrate the data now lags far behind our data-collection capabilities [34]. Source-based ammonia emission data from a limited number and different types of studies have been compiled to build inventories thus far. This technique can lead to large errors and inconsistencies. In addition, most of the ammonia emissions inventories published by U.S. Environmental Protection Agency (EPA) [17] are based on European research [35].

As a consequence, while the knowledge and models needed for agricultural ammonia emission inventories are still very limited, comprehensive field measurement data are generally far from being fully exploited and rich information containing insights into the knowledge about unknown factors governing pollution generation, release, and emission wait to be analyzed and used to improve emission inventories and control strategies. Research is therefore urgently needed to bridge the gap.

2.2. Emission Modeling

Ammonia emission factors are widely used for modeling regional and national ammonia emissions to obtain emission inventories such as those in the U.S. [17] and in Europe [36]. The concept of the emission factor is simple to apply and is usually based on a few input variables including type and number of animals and manure management systems. It adds the emissions from each farm element to obtain regional and national ammonia emission inventories. Ammonia emission from animal buildings is related to animal species, diet and excreta composition, distribution of emitting surfaces, mass transfer characteristics, quantity, composition, and management of manure, as well as environmental variables [37]. The National Research Council [38] proposed replacing the “emissions factor” approach with a “process-based modeling” approach. The process-based model incorporates “mass balance” constraints for some of the emitted substances of concern and uses a mathematical model to represent the interactions between the system components. The National Research Council committee recommended using process-based modeling to predict emissions from both individual AFOs and regions. The committee claimed that, if pursued vigorously, process-based modeling can enhance both regulation and management of air pollution emissions. Furthermore, a process-based model farm approach, in conjunction with estimated emission factors for other substances, may be a useful alternative to the EPA model farm concept.

Several studies have been published on process-based modeling in attempts to estimate ammonia from animal agriculture [39-44]. Zhang *et al.* [45] developed the most comprehensive process-based model ever published in a science document, in which details of a Farm Emission Model (FEM) for ammonia emissions from AFOs were described. The model was based on new concepts and sub-models, as well as existing theories and models published in the scientific literature, including the work of Pinder *et al.* [40]. The document established a conceptual framework of the FEM that contained four sub-models for animal excretion, confinement housing emission, feedlot emission, and land application emission. However, this latest process-based ammonia model still has several shortfalls:

1. The process-based model may lack accuracies because coefficients and parameters in some sub-models (e.g., mass transfer sub-model) obtained in lab conditions may be inapplicable in field conditions.

2. The amount of variations that the model can explain with current scientific understanding is limited and all of the experimental results to date cover only a small subset of possible emission scenarios [40]. In addition, most of the sub-model parameters were based on various studies conducted in Europe in the 1980s and 1990s that lack consistency and are not up to date. Some of them just use simplified assumptions or constants.
3. The model and sub-models were not validated individually using field research data [45].
4. The gas release process is very complex with abundant nonlinear relationships between gaseous emissions and the many variables that cause gas production [46]. Some circumstances of gas production, release, and emission are not well understood and model parameters are very difficult to determine experimentally under field conditions.
5. The application of the models presents practical difficulties because they require large amount of inputs, of which many are tricky to obtain at farm levels, especially for large scale applications.
6. Understanding of all details of biological, chemical, and physical processes involved in ammonia emission is impossible in the near future. Therefore, submodels of process-based ammonia emission models have to be developed empirically. Compared with emission factors, process-based modeling depends less on empirical approaches and more on complex methodology.

It becomes obvious that, while the emission factor is a simplified modeling approach that needs enhancement, the current process-based model approach needs substantial improvement before it can become an accurate, practical, and user-friendly tool for emission inventories.

2.3. Connection between Data and Modeling Tools

Scientific research to connect the experimentally obtained field monitoring data as shown in Table 1 and the modeling tools is an urgent task in agricultural ammonia emission regulations and mitigation. The gaps between data and modeling tools can be filled by the new knowledge obtained from the data and applied in the emission models to enhance the accuracy of national ammonia emission inventories. Novel approaches to analyze the data are necessary because the size and complexity of the latest ammonia monitoring database are unprecedented. The hypotheses of this scientific endeavor are:

1. With sufficiently large datasets of high quality, new knowledge in physical processes and important relationships affecting ammonia emissions and variations can be discovered. One or more new factors or processes that affect ammonia emission will benefit the overall scientific community.
2. The new knowledge can be used to significantly improve existing emission factors and process-based models by assessing them to identify known and suspected errors, estimating fundamental model parameters with measurement data, and validating the models to increase model accuracy and applicability, and optimizing the model structures to the key factors/processes affecting emissions.
3. The improved modeling tools can be applied at regional and national level ammonia emission calculations for current and near-term future ammonia emissions with high

temporal resolution (at hourly, daily, or weekly scales) from animal housing and the potentials for emission reductions.

3. Materials and Methods

3.1. The NAEMS Database

The National Air Emissions Monitoring Study (NAEMS) was initiated to improve the emission database. The NAEMS has generated a unique database that can greatly contribute to the advances in understanding agricultural air pollution and improving the accuracy of emission inventories (Table 2).

Table 2. Overview of 14 NAEMS sites in eight states in U.S. and the data acquired.

State[a]	Building type (year) [b]	#Building	#Head/building	#SAS[c]	#Variable
Broilers: >131M data points					
CA	TMV, litter on floor (2002)	2	21,000	7 (7)	125
Layers: >771M data points					
CA	MV, high-rise, DB (2003)	2	38,000	7 (7)	104
IN	MV, high rise, CBC (1997)	2	250,000	15 (15)	169
	MV, manure belt (2004)	2	280,000	13 (19)	321
	NV, manure shed (2004)	1			
NC	MV, high rise, CBC (2003)	2	103,000	7 (7)	140
Swine: >635M data points					
IN	TMV, finishing, deep pit (2003)	4	1,000	17 (17)	154
NC	TMV, finishing, PPR (1995)	3	800	4 (4)	98
IA	TMV, gestation, deep pit (1998)	2	1,100	12 (18)	147
	MV, farrowing, PPR (1998)	1	24		
NC	TMV, gestation, PPR (1994)	2	850	6 (6)	85
	MV, farrowing, PPR (1995)	1	24		
OK	TMV, gestation, PPR (1994)	2	1,200	12 (12)	121
	MV, farrowing, PPR (1994)	1	24		
Dairy: >911M data points					
CA	NV, freestall, flushing (2001)	2	600	11 (45)	239
IN	TMV, freestall, scrape (2004)	2	1,600	17 (23)	186
	MV, milking center, flushing (2004)	1	562		
NY	TMV, freestall, scrape (1998)	1	470	7 (7)	107
	MV, milking center, deep pit (1990)	1	187		
WA	NV, freestall, flush (2002)	2	650	11 (33)	214
WI	MV, freestall, scrape (2007)	2	325	11 (11)	121
	MV, milking center (1990)	1	80		

Note: Adapted from Heber *et al.* [33]. [a] State where the monitoring site was located. [b] Year = year of construction, PPR = pull-plug with recharge, DB = dropping board, CBC = curtain-backed cages, MV = mechanically ventilated, NV = naturally ventilated, TMV = tunnel mechanically ventilated. [c] SAS = sample air streams provided to gas analyzers. Numbers in parentheses are sample locations. Some air streams were from multiple air sampling locations.

The NAEMS was the world's largest air pollution monitoring campaign for animal agriculture. Its barn-component covered 35 mechanical and naturally-ventilated swine, dairy, layer, and broiler buildings and one naturally ventilated layer manure shed at 14 farms in eight states that were representative of the AFOs in the U.S. The total cost of obtaining these data was about \$15M.

Ammonia concentrations in the NAEMS were determined by sampling air at selected gas sampling locations in the building air inlets and exhausts using photoacoustic Field Gas Monitors (Model 1412, Innova AirTech Instruments, Ballerup, Denmark) [33]. Airflow rates were calculated based on ventilation fan monitoring and building static pressure measurement for mechanically-ventilated buildings. For naturally-ventilated buildings, they were calculated based on wind velocity measurement at building walls and ridge openings. Ammonia emission rate was calculated as the product of ammonia concentration difference between building inlet and exhaust and building airflow rate. The NAEMS also monitored emissions of other aerial pollutants, including hydrogen sulfide, carbon dioxide, methane, volatile organic compounds (VOC), and particulate matter. Additionally, the NAEMS monitored gases from area sources, mainly from manure storage lagoons. Information about farm operations, including feed input, animal growth or milk/egg production, manure generation and composition, *etc.*, was also collected.

However, the scope of NAEMS did not cover aerial emissions from manure applications onto farm fields and from the feed storages on production operations. The scope of this paper does not include aerial pollutants besides ammonia.

The NAEMS database had the following attributes:

1. The number of measurement data points (each was the measurement of one variable for 1 min) in the NAEMS database was 2.4 billion. It was 2.5 times as many as the sum of the 0.9 billion data points collected by all previous long-term (>6 months) and continuous on-farm monitoring projects in the U.S.
2. A standardized monitoring protocol was used at all sites to generate the most consistent multi-farm air quality monitoring data.
3. The highest level quality assurance and quality control (QAQC) plan, approved by the U.S. EPA, was developed and applied [47]. The U.S. EPA's Office of Air Quality Planning and Standards (OAQPS) in Raleigh, North Carolina provided QAQC oversight throughout the study.
4. The two-year continuous monitoring period was the longest among all similar projects. The 24-month period makes it possible to analyze annual emission variations for the first time.
5. The farm production process inputs and outputs, including animal inventories, feed and water consumption, meat, milk, egg, and manure production, and building management, were recorded or sampled to allow mass balance studies.

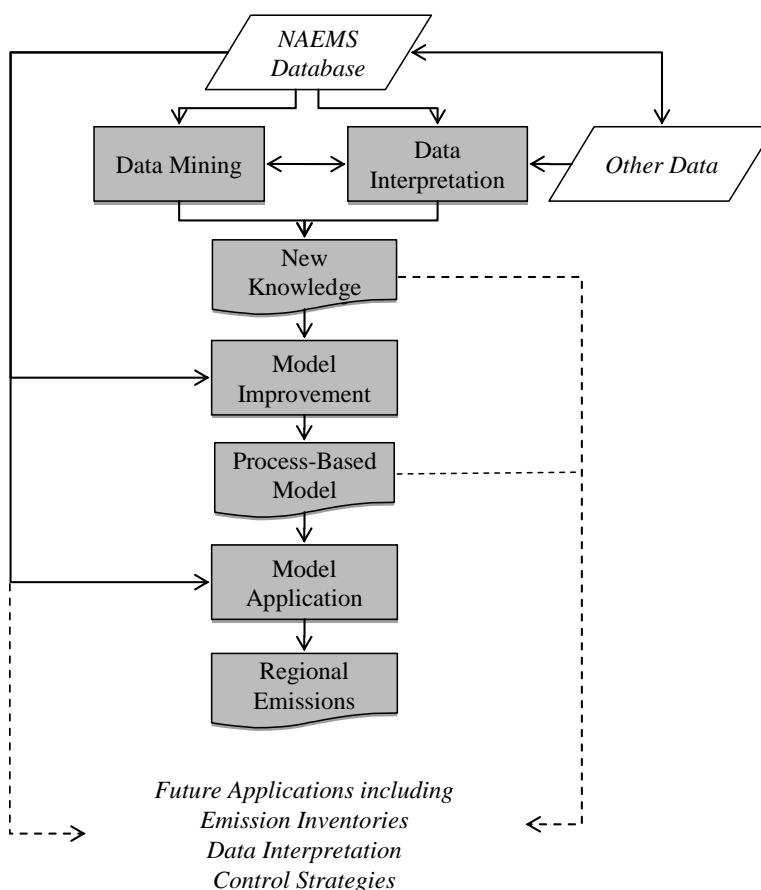
The original NAEMS database end-use plan included analyzing the data using a pre-defined data processing procedure to obtain emission rates that were submitted to the U.S. EPA OAQPS in 2010 for their development of "emission estimating methodologies." The only data submission requirement was the simple hourly and daily means of the data. The NAEMS stopped short of characterizing and interpreting the database, leaving much to be learned from it.

The NAEMS database deserves more intensive study by qualified personnel, who not only have the expertise for data analysis and modeling, but also a familiarity and knowledge about the data and how the data were collected, to bridge the gap between national emission monitoring data and national ammonia emission inventories, using novel approaches.

3.2. General Approach

Figure 1 illustrates the general approach and activities in this work, as well as its future applications. The NAEMS database will be used to obtain new knowledge via novel approaches of data mining and intelligent interpretation. Data mining will help to uncover otherwise hidden patterns about ammonia emission in the database and intelligent data interpretation will help to determine the physical laws behind these patterns.

Figure 1. Diagram of the general research approaches, activities, results, and applications.



The new knowledge will be used to characterize and correct errors, explain outliers, determine bounds of uncertainty and confidence, and delineate short-falls in the current emission inventories. It will be applied to refine and improve current process-based models for ammonia emission from animal housing. The on-farm measurement data will be used to estimate model parameters, and test and validate the model.

Selected data from previous studies will be used to compare with the NAEMS data and to accept or reject the new knowledge obtained from NAEMS study. The NAEMS data and the new knowledge

will be used to develop improved modeling techniques, which will be tested and applied to calculate the current and near-term regional and national ammonia emissions.

3.3. Data Mining

Data mining, or KDD (Knowledge Discovery in Databases), depends on non-trivial extraction of implicit, previously unknown, and potentially useful knowledge from large amounts of data. It is a hybrid discipline that integrates technologies of databases, statistics, machine learning, signal processing, and high performance computing [48]. It has been increasingly used in the areas of science and engineering, such as bioinformatics, genetics, medicine, education, and electric power engineering in recent years. Data mining techniques that have been successfully developed and applied in different research areas include classification and clustering, time-series analysis, classification using decision trees, support vector machines kernels, association discovery, and detection of sequential patterns [34]. Application of data mining in air pollution has also started and results have been obtained [34,48-50]. However, data mining has not yet been used for discovering new knowledge from agriculture-specific air pollution databases.

The results or rules generated by data mining are empirical—they are not physical laws. However, while there are no general theories, data mining techniques are valuable, especially where one has large quantities of data containing noisy patterns [51]. Therefore, data mining is a new tool that is valuable in a learning process for analyzing 2.4 billion data points generated by the world's largest field testing and sampling of AFO air pollution. The potential new knowledge revealed by data mining can provide insight into the complex ammonia emission problem and improve predictive emission models and ammonia inventories.

Data mining will be applied in the “training set” and “test set” data from the NAEMS database (described later in this section). Algorithms such as artificial neural networks will be tested to develop applicable data mining method. The method performance will be verified with the “test set” data. Refinement of data mining methods previously used in other applications is expected.

The data mining technique will be employed for potential new knowledge discovery to increase understanding of ammonia emission from animal housing systems. It will begin with data from one of the NAEMS monitoring sites for method development in relation to agricultural air quality data, and gradually expand its application during the learning process as experiences are obtained and methods are established. Custom data processing software, CAPECAB [52,53], and commercial data mining software, e.g., SAS[®], will be utilized.

Data mining of the NAEMS database will provide the first insights into the data. This stage of work will be based on the familiarity of the data and the test farms will focus on some new questions raised during field monitoring and regular data processing. Although data mining does not test or apply specific process-based calculations, the technique will help to uncover existing data patterns, and with data interpretation and characterization to be described in the next section, to get answers to some specific questions:

1. Why was there significant variability in ammonia emissions from paired and almost-identical animal buildings at the same farm, as identified during NAEMS monitoring?

2. Were there annual variations in ammonia emission from the same emission sources and what were their magnitudes?
3. What were the geographical variations of ammonia emissions at similar farms using identical monitoring protocols?
4. Did animal and worker activities affect ammonia emission significantly?
5. What was the relationship between animal age and ammonia emissions?
6. Was there significant interaction between the emissions of ammonia and emissions of other gaseous pollutants, including hydrogen sulfide and carbon dioxide, which were also monitored in the NAEMS, under field conditions?
7. Were there special temporal patterns of ammonia emissions related to the weather (e.g., consecutive rainy days) and farm management (e.g., manure removal from manure belt buildings according to day of the week)?
8. What was the dynamic effect of ventilation control strategies on indoor ammonia concentrations?
9. What were the main causes of spatial variations of ammonia concentration/production in animal buildings? It was observed at NAEMS sites that ammonia production at some locations could be as much as 10 times at other locations in the same building, especially during low ventilation periods in winter.
10. Was the spatial variation of ammonia release from manure in under-floor storage pits a fundamental obstacle when using the mass transfer sub-model in process-based ammonia emission models?
11. Did water vapor content have an effect on ammonia emissions?
12. Were there ceilings of ammonia emissions under the worst-case scenario for building conditions (e.g., full manure storage, maximum animal capacity, high temperature, high ventilation, *etc.*) that can be used as boundaries in predictive ammonia emission modeling?

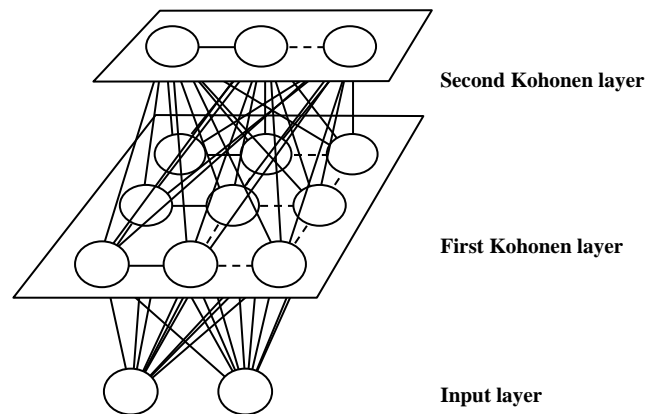
Although there is no general framework for systematically applying analysis techniques in a multi-step data mining process, and much of the time involved in data mining efforts is the user-driven, interactive, exploratory algorithms [34], a 4-stage data mining process that includes data understanding, data preparation, modeling, and result interpretation will be undertaken in this study [48,51].

In the data preparation stage, selected data that are large enough to contain uncovered patterns while remaining concise enough to be mined in an acceptable timeframe will be assembled for a target dataset. The data will be reduced into feature vectors, one vector per observation. A feature vector is a summarized version of the raw data observation that dramatically reduces the size of the dataset to be mined, and hence reduces the processing effort. Selection of the “right” feature(s) is fundamental to successful data mining and will be based on the ammonia emission monitoring and modeling experience, and data and farm familiarity of the research team. The feature vectors will be divided into two sets. A “training set” will be used to “train” the data mining algorithms. A “test set” will be used to verify the accuracy of any patterns found.

In the modeling stage, common algorithms, such as artificial neural networks [46,48], will be used for association rule learning and searching for relationships among variables. A two-layer neural network topology [48,54] will be adapted (Figure 2). Regression will be run to find functions which

model the data or data subsets with the least error. Algorithms other than the artificial neural network as described in [34] will also be tested to obtain most satisfactory results.

Figure 2. The architecture of two-layer neural network for air quality data mining. Reprinted from [48], with permission from Elsevier.



In the result interpretation stage, patterns produced by the data mining algorithms will be evaluated. To overcome the “over-fitting” problem during data mining, the “test set” of data will not include any data used in the training procedure. The learned patterns will be applied to the "test set" and the resulting output will be compared with the desired output. If the learned patterns do not meet the desired standards, then the data understanding, data preparation and/or modeling steps taken will be reevaluated and changed, if necessary. If the learned patterns meet the desired standards, then the final step is to interpret the learned patterns and turn them into knowledge. The resultant theory, while maybe not fundamental, can yield a good understanding of the physical process and can have great practical utility [51].

3.4. Data Interpretation

Intelligent interpretation of ammonia emissions will be based on measurement data from the NAEMS database and other selected major monitoring studies, in parallel with the data mining procedure. Published ammonia emission and emission inventories at different times, using different methods, and from different geographical locations, including Europe and North America, will be collected and evaluated. The data from two to three comprehensive ammonia monitoring projects that are suitable for comparison with the NAEMS data will be selected. The selection will emphasize representativeness, amount of available information, and quality of data to ensure that a critical check of the data will yield meaningful results.

The background of the NAEMS and other data used in this study, including how the data were obtained and calculated and the uncertainties, will be critically inspected. Physical and chemical laws, statistical analyses, existing and new knowledge about ammonia emissions, comparison with scientific publications, and verification with standards and guidelines (e.g., manure production, farm management, *etc.*) will be used to interpret the emission data. The interpretation will focus on answering the following questions:

1. What were the major causes of the discrepancies among ammonia emission factors (e.g., methodology and technology in on-farm monitoring, monitoring durations, geographical location of the monitoring sites, data processing and emission calculation, unusual or infrequent farm operations, *etc.*)?
2. Were the major causes explainable with existing and new knowledge about ammonia emission from animal housing?
3. What uncertainties can these major causes introduce into the emission inventories?
4. How much can the NAEMS data contribute to the improvement of national ammonia inventories?
5. What are the pitfalls related to the monitoring data (e.g., quality, quantity, and representativeness of the data, *etc.*) and the way the data are used?
6. What critical information is missing from the data for emission modeling and inventories that need to be addressed in future on-farm monitoring?
7. What recommendations can be made to improve future on-farm data collection? Will standardization of the methodology and technology in data collection and processing greatly improve the accuracies of emission inventories?
8. What recommendations can be obtained from the monitoring data to help develop emission control strategies, especially best management practices?

Data interpretation is expected to further improve understanding of ammonia emission from animal housing systems. The new knowledge obtained in this study with the NAEMS will be verified with other datasets. Whether the knowledge is generally applicable will be confirmed or rejected. Additionally, the interpretation of the datasets will help to improve the accuracy of emission inventories and emission prediction modeling.

3.5. Reduced Process-Based Modeling

The confinement housing ammonia emission algorithms proposed by Sommer *et al.* [37] and in a sub-model of the FEM developed by Zhang *et al.* [43] will be assessed using four sets of input variables available from selected NAEMS sites, one set from each animal species: swine, dairy, layers, and broilers. Data with different temporal resolutions, *i.e.*, every min, hour, day, and month will be tested. The objective of assessment is to determine whether the model is of sufficient quality to inform a regulatory decision.

The model assessment results will be compared with real on-farm system data to identify the causes of significant discrepancies. Assessments will follow the U.S. EPA guidance on model evaluation [55], underline the model's merits and pitfalls, and focus on the following factors: (a) How have the principles of sound science been addressed in the model? (b) How is the choice of model supported by the quantity and quality of available data? (c) How closely does the model approximate the real system? (d) How well does the model perform the specified task while meeting the model objectives?

Improvement of the current process-based models will be focused on the following to increase the model accuracy, robustness, and applicability:

1. Determine the optimal level of model complexity by making appropriate tradeoffs among competing objectives.

2. Reducing the 80–100 model variables by combining or removing less important model inputs that have negligible effects on model output and also are difficult or impossible to obtain during large scale application of the model at farm level (e.g., the length, width, and height of the buildings, weight of dairy cows, *etc.*).
3. Characterize model parameters with direct on-farm measurement data, combined with insights obtained during data mining whenever possible, rather than relying on assumptions or simplified constants, and use input data that meet data quality acceptance criteria.
4. Revise or develop sub-model(s) according to the new knowledge and theories obtained during data mining and emission interpretation.
5. Study the possibility of replacing some highly non-linear and complex mechanistic sub-models [46] (e.g., the mass transfer sub-model that is extremely sensitive to the model output yet difficult to determine accurately in field conditions) with sub-models of reduced structure.
6. Develop functions for negative feedback and emission boundaries learned during data mining in the model.

Sensitivity and uncertainty analyses will be performed during several model development stages using the global method with Monte Carlo simulation [56]. Sensitivity analysis evaluates the effect of changes in input values or assumptions on a model's results to identify the most important ones that could assert large influences on the overall ammonia emission rates from animal housing systems. Uncertainty analysis investigates the effects of “lack of knowledge” and other potential sources of error in the model (e.g., the “uncertainty” associated with model parameter values). When conducted in combination with sensitivity analysis, uncertainty analysis allows a model user to be more informed about the confidence that can be placed in model results [55].

The model improvement process will be conducted for one animal species a time and tested with part of the on-farm data that is large enough to expose potential problems in the model for follow-up model fine-tuning. The final version of the improved model will be validated following the standard guide of ASTM [57] using data that have not been applied in model development.

3.6. Model Testing for Regional Emission Inventory Calculation

The improved model will be used to calculate ammonia emissions at the farm level for swine, dairy, and poultry housing systems in selected regions in the U.S. The model will be run to generate distributions of animal housing ammonia sources at fine temporal and spatial resolutions of hourly and daily time scales and at farm levels to identify and quantify the amount, location, and timing of ammonia emissions. In addition, several scenarios with predicted near-future animal agriculture development and potential application of ammonia control technologies will be developed and their effects on ammonia emissions will be calculated.

Model results will be verified by randomly selecting a sample of 30 farms and comparing the inputs and outputs of the model calculation from each farm with comparable measurements from the NAEMS and other studies. If significant and unexplained discrepancies are identified, the causes of the difference will be studied and the calculation performed again. The monthly mean ammonia emissions

at all farms of the same animal species in each state will also be compared with other published emission data, taking into account the differences in geography, farm management, *etc.* The modeling results will be mapped to display temporal variations and geographical distributions of the ammonia emission density.

3.7. Model Refinement and Validation

The tested and improved model(s) will be evaluated for magnitude and time-dependant response. Initial model evaluation will be based on the Standard Guide for Statistical Evaluation of Indoor Air Quality Models [57]. This guide provides multiple tools and suggested limits for assessing model accuracy. An important component of model testing described in ASTM [57] and in other model literature [e.g., 58-60] is the selection of independent data for model evaluation from the data used for model development and testing. Validation data will be extracted prior to sub-model development and will not be used in calibration procedures. The refined and validated model will be used to expand its application to national level ammonia emission calculations.

4. Expected Results

The outputs of this study will enhance science and research in agricultural air quality. It will provide valuable feedback for the improvement of ammonia emission monitoring methodology and technology, and help to derive best management practices to reduce ammonia emissions. Methods developed in this study can also be applied to the analysis and modeling of inventories of other gaseous or aerosol pollutants, as well as at other agricultural air pollution sources such as open feedlots, manure treatment lagoons, and field application of animal wastes.

The new techniques of data mining can uncover hidden patterns in a large database and intelligent interpretation can find explanations of physical laws behind the new factors affecting ammonia generation and emission. The new knowledge, methodologies, algorithms, and models developed in this research can help future characterizations and reduce known and suspected errors and other shortfalls in the emission inventories.

The refined process-based emission models will generate ammonia emission source data with improved accuracy and finer temporal resolutions (every min, hourly, or daily). The data can be used as inputs to air quality models to represent the initial introduction of air pollutants into the atmosphere. Because ammonia is an important contributor to $PM_{2.5}$ mass in some places, the reliable and location-specific ammonia emission data can also help to understand atmospheric chemistry and/or physics under the changing conditions due to implementation of major emission reductions, regulations, or rules.

5. Summary and Conclusions

Although agricultural ammonia emission has been researched for more than five decades, the current ammonia emission inventories are still highly uncertain and the modeling tools need fundamental improvement. This was partly because of a significant disconnection between the field monitoring data and ammonia emission inventories.

There have been ten long-term (>6 months) and continuous field measurements monitoring at a total of 78 animal buildings in 12 states in the U.S. that have produced 3.3 billion comprehensive field sampling data points. However, integration of these data into regional and national emission inventories lags far behind our data-collection capabilities. We not only need comprehensive data but, more importantly, the knowledge about the data we have.

The current situation can be greatly improved by bridging the gap between the data and application of the knowledge from the data into modeling tools and emission inventories. New research ideas and approaches are proposed to explore the data from the NAEMS and other comprehensive datasets using data mining and intelligent interpretation.

A series of questions about the characteristics of ammonia emission that were raised from field experiments are expected to be answered. The fundamental understanding of ammonia emissions from U.S. agriculture will be enhanced and the accuracy and scope of regional and national ammonia emission inventories will be significantly improved if the gap is filled.

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