Sonoma Technology, Inc.

1360 Redwood Way, Suite C
Petaluma, CA 94954-1169
707/665-9900
FAX 707/665-9800
www.sonomatech.com

RECOMMENDED IMPROVEMENTS TO THE CMU AMMONIA EMISSION INVENTORY MODEL FOR USE BY LADCO

FINAL REPORT

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By:
Lyle R. Chinkin
Patrick A. Ryan
Dana L. Coe
Sonoma Technology, Inc.
1360 Redwood Way, Suite C
Petaluma, CA 94954

Prepared for:
Michael Koerber
Lake Michigan Air Directors Consortium
2250 East Devon Ave., Suite 250
Des Plaines, IL 60018

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TABLE OF CONTENTS

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LIST OF FIGURES

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LIST OF TABLES

EXECUTIVE SUMMARY

This study was sponsored by the Lake Michigan Air Directors' Consortium (LADCO) to provide guidance and recommendations for revisions that improve the performance of the Carnegie Mellon University (CMU) ammonia emissions model for the LADCO region. Highly resolved ammonia emission inventories are needed to address the U.S. Environmental Protection Agency's (EPA) regional haze and particulate matter (PM) regulations. To help address these needs, Strader et al. (2002a) of CMU developed an ammonia emissions modeling tool. The CMU model outputs ammonia emissions for numerous source categories for each county of the United States. However, it is the responsibility of model users to evaluate the model algorithms and pre-loaded databases, to determine whether they are best for use in the model users' geographic areas of interest, and to apply updates and improvements as needed. The goals of this study are to assist LADCO with an evaluation of the CMU model, to mine existing sources of information, and to recommend revisions as necessary.

SUMMARY OF RECOMMENDATIONS

Table ES-1 provides a summary of recommended emission factors for comparison with those that are currently used in the CMU model. **Tables ES-2 and ES-3** list our recommendations for temporal allocation profiles. The net effect of adopting all of STI's recommendations is to decrease total estimated ammonia emissions for the LADCO and surrounding states by about 15% (ignoring biogenic soil emissions). While livestock is still the most significant source of ammonia emissions, its significance declines from about 65% to 55% of total emissions. Fertilizer emissions remain the second most significant source, and their contribution to the total inventory increases from about 20% to 30%. Mobile source emissions become a significant, though small, source (about 5% of the inventory) while the contribution from publicly owned treatment works (POTWs) fell from 3% to a negligible amount, reflecting the recommended ammonia emission factor.

Table ES-1. Recommended ammonia emission factors for comparison with factors used in the CMU model.

Source Category	CMU Model	Recommendation	Units
On-road motor vehicles			
Gasoline-powered vehicles	***0.013455***	0.097[a]	g NH_3/VMT
Trucks (diesel-powered)	***0.068262***	0.027	g NH_3/VMT
Off-road mobile sources			
Gasoline-powered	-	0.15	g NH_3/gallon
Diesel-powered	-	0.17	g NH_3/gallon
Waste Treatment			
POTWs	*8.300*	0.054	kg $NH_3/10^6$ gallons
Landfills	-	0.007	kg NH_3/kg methane emitted
Livestock			
Milk cows	***39.720***	25.00	kg NH_3/cow/yr
Beef cattle	***39.720***	9.00	kg NH_3/cow/yr
Hogs and pigs	***9.200***	7.00	kg NH_3/pig/yr
Poultry	***0.167***	0.22	kg NH_3/poultry/yr
Horses	***12.200***	5.10	kg NH_3/horse/yr
Sheep	***3.370***	0.46	kg NH_3/sheep/yr
Goats	***6.400***	0.46	kg NH_3/goat/yr
Fertilizer			
Mix	4.0	7.0	% of applied nitrogen content
Anhydrous ammonia	***1.0***	4.0	% of applied nitrogen content
Aqueous ammonia	***1.0***	4.0	% of applied nitrogen content
Ammonium nitrate	***2.0***	1.0	% of applied nitrogen content
Ammonium sulfate	***8.0***	5.0	% of applied nitrogen content
Ammonium thiosulfate	2.5	2.5	% of applied nitrogen content
Calcium ammonium nitrate	***2.0***	1.0	% of applied nitrogen content
Nitrogen solutions	8.0	8.0	% of applied nitrogen content
Urea	15.0	15.0	% of applied nitrogen content
Diammonium phosphate	***4.0***	5.0	% of applied nitrogen content
Monoammonium phosphate	***4.0***	5.0	% of applied nitrogen content
Liquid ammonium polyphosphate	***4.0***	5.0	% of applied nitrogen content
Potassium nitrate	***2.0***	1.0	% of applied nitrogen content
Miscellaneous	***15.0***	7.0	% of applied nitrogen content

[a] Equivalent to approximately 8% of NO_x emissions.

Table ES-1. Recommended ammonia emission factors for comparison with factors used in the CMU model.

Source Category	CMU Model	Recommendation	Units
Biogenic Soils			
Bare soil	***365***	370	kg/km^2-yr
Cropland	***1241***	0.0	kg/km^2-yr
Desert scrub	***365***	6.7	kg/km^2-yr
Grassland	***365***	40	kg/km^2-yr
Pasture	***1241***	550	kg/km^2-yr
Rangeland	***365***	14	kg/km^2-yr
Scrubland	***365***	100	kg/km^2-yr
Urban land area	***160.6***	400	kg/km^2-yr
Lawn surface	***160.6***	370	kg/km^2-yr
Oak forest	***365***	1.6	kg/km^2-yr
Pine forest	***365***	1.1	kg/km^2-yr
Other coniferous forests	***365***	40	kg/km^2-yr
Temperate forest and woodland and shrubland	***365***	400	kg/km^2-yr

Table ES-2. Recommended seasonal profiles.

Source Category	Season			
	Winter	Spring	Summer	Fall
Mobile Sources	25%	25%	25%	25%
POTWs	25%	25%	25%	25%
Cattle and Calves	Use the adjusted Gilliland et al. (2002) monthly profile shown in Table 3-2.			
Pigs and Hogs				
Other Livestock				
Fertilizer	Use the seasonal allocation pre-loaded in the CMU ammonia model			

Table ES-3. Recommended diurnal profiles.

Hour of Day	Mobile Sources	POTWS	Cattle and Calves	Pigs and Hogs	Other Livestock	Fertilizer
Midnight-1 a.m.	1.1%	4.2%	3.9%	3.9%	3.9%	2.0%
1-2 a.m.	0.7%	4.2%	4.0%	4.0%	4.0%	2.0%
2-3 a.m.	0.5%	4.2%	4.0%	4.0%	4.0%	2.0%
3-4 a.m.	0.6%	4.2%	4.1%	4.1%	4.1%	2.0%
4-5 a.m.	0.9%	4.2%	4.1%	4.1%	4.1%	2.0%
5-6 a.m.	1.7%	4.2%	4.2%	4.2%	4.2%	2.1%
6-7 a.m.	3.3%	4.2%	4.2%	4.2%	4.2%	2.8%
7-8 a.m.	4.9%	4.2%	4.2%	4.2%	4.2%	4.1%
8-9 a.m.	5.4%	4.2%	4.2%	4.2%	4.2%	7.0%
9-10 a.m.	4.9%	4.2%	4.3%	4.3%	4.3%	7.4%
10-11 a.m.	4.8%	4.2%	4.3%	4.3%	4.3%	8.2%
11 a.m.-Noon	4.8%	4.2%	4.3%	4.3%	4.3%	8.2%
Noon -1 p.m.	4.9%	4.2%	4.3%	4.3%	4.3%	8.1%
1-2 p.m.	5.5%	4.2%	4.3%	4.3%	4.3%	7.8%
2-3 p.m.	6.2%	4.2%	4.3%	4.3%	4.3%	6.5%
3-4 p.m.	6.5%	4.2%	4.3%	4.3%	4.3%	4.1%
4-5 p.m.	6.8%	4.2%	4.2%	4.2%	4.2%	4.1%
5-6 p.m.	7.5%	4.2%	4.2%	4.2%	4.2%	3.1%
6-7 p.m.	7.4%	4.2%	4.2%	4.2%	4.2%	2.9%
7-8 p.m.	6.7%	4.2%	4.2%	4.2%	4.2%	2.9%
8-9 p.m.	5.7%	4.2%	4.1%	4.1%	4.1%	2.9%
9-10 p.m.	4.5%	4.2%	4.1%	4.1%	4.1%	2.9%
10-11 p.m.	2.9%	4.2%	4.0%	4.0%	4.0%	2.9%
11 p.m.-Midnight	1.8%	4.2%	4.0%	4.0%	4.0%	2.0%

1. INTRODUCTION

Highly resolved ammonia emission inventories are needed by the Lake Michigan Air Directors' Consortium (LADCO) and regional planning organizations (RPOs) to address the U.S. Environmental Protection Agency's (EPA) regional haze and particulate matter (PM) regulations. To help address these needs, Strader et al. (2002a) of Carnegie Mellon University (CMU) developed an ammonia emissions modeling tool. The CMU model outputs ammonia emissions for numerous source categories for each county of the United States. It is a framework that incorporates activity data, emission factors, and temporal information. In addition, it includes pre-loaded databases of activity parameters, emission factors, and, for one source category, fertilizer consumption, monthly activity levels. It distributes emissions spatially to the county level and is capable of producing gridded outputs of county-level emissions.

It is the responsibility of model users to evaluate the model algorithms and pre-loaded databases, to determine whether they are best for use in the model users' geographic areas of interest, and to apply updates and improvements as needed. Sonoma Technology, Inc.'s (STI) goals are to assist LADCO with an evaluation of the CMU model, to mine existing sources of information, and to recommend revisions that improve the performance of the CMU model for the LADCO region. Our recommendations, presented in this report, concentrate primarily on immediately available revisions and secondarily on long-range research goals.

1.1 SOURCES OF AMMONIA AND THEIR SIGNIFICANCE

Ammonia is one of the precursors to secondary PM. Other precursors are nitrogen oxides (NO_x), sulfur oxides (SO_x), and volatile organic compounds (VOCs). Ammonia (NH_3) contributes to the formation of secondary PM through a series of gas- and aqueous-phase chemical reactions with NO_x or SO_x. The reaction products are ammonium nitrate and ammonium sulfate, both of which promote and stabilize the formation of very tiny particles—particles that very effectively penetrate lung tissues and degrade visibilities. Ammonium sulfate contributes substantially to secondary particle mass measurements in urban areas of the eastern United States. In the western United States, ammonium nitrate plays the larger role. Because ammonia plays such a key role throughout the Unites States in the formation of secondary PM, accurate inventories of ammonia emissions are crucial to the development of realistic air quality modeling results and effective regional haze and PM control strategies.

Current uncertainties in ammonia emission inventories are much greater than in emission inventories of primary pollutants, such as SO_x and NO_x. Primary emissions of SO_x and NO_x have a long history of scrutiny because of their immediate health effects and roles in acid rain and ozone formation. Additionally, major point sources, which are relatively easy to monitor, are significant contributors to SO_x and NO_x emissions. In contrast, ammonia is associated with an array of rural and urban source types (listed below), many of which are diffuse or unregulated.

Rural Activities

- Decomposition of livestock and poultry wastes
- Natural biological cycling (due to biotic processes in soils and waters)

- Fertilizer application
- Landfills
- Composting
- Geothermal emissions
- Combustion – biomass (forest fires and agricultural fires)

Urban Activities

- Mobile sources
- Wastewater treatment plants (including sewage sludge)
- Combustion – industrial, commercial, and residential
- Nitrogenous materials manufacturing (fertilizers, etc.)
- Fossil fuels processing (coke production, catalytic cracking)
- Ammonia injection as a control measure (power generation plants)
- Ammonia refrigeration
- Domestic sources (solvent use, cleaners, untreated wastes, etc.)
- Commercial Ammonia use (printing processes–blueprints, solvents, cleaners, etc.)

Despite this lengthy list, a relatively small number of source categories account for the majority of ammonia emissions. Rural sources—including decomposition of livestock and poultry wastes, natural biological cycles in soils, and fertilizer application—dominate the total mass of ammonia emissions across large spatial scales. The EPA's emission inventory indicates that livestock management and fertilizer application contributed about 85% of total ammonia emissions in the United States in 1998, while publicly owned treatment works (POTWs), mobile sources, and combustion sources contributed only about 15% of the total (U.S. Environmental Protection Agency, 2002b). However, we cannot rely entirely on national magnitudes to determine the relative importance of emissions from POTWs, mobile sources, and combustion sources. On smaller scales, in urban areas, or during certain time periods, these sources can influence the inventory more significantly. In many urban areas of the Unites States, for example, we believe that on-road mobile sources contribute significantly to ammonia emissions, especially during the wintertime when freezing temperatures slow down bacterial activity in soils and organic wastes. The relative increase in importance of these more urban sources is partly a result of the formation of secondary nitrate aerosols in the atmosphere that requires the coexistence of ammonia and either NO_x or SO_x. Therefore, the accuracies of ammonia emissions estimates for sources that (a) co-emit SO_x or NO_x and/or (b) emit into ambient plumes of SO_x or NO_x are much more important than their absolute magnitudes might suggest. Although emissions from livestock and fertilizer application are large in magnitude, they are mostly geographically removed from sources of NO_x or SO_x. Conversely, although emissions from POTWs, mobile sources, and combustion sources are small, they are usually closely associated with NO_x or SO_x emissions.

1.2 NEW AND ONGOING RESEARCH PROJECTS

While conducting our evaluation of the CMU model, we also sought to avoid duplicating others' on-going efforts. In particular, the LADCO requested that we consider three recent studies:

- National Academy of Sciences' report entitled "Air Emissions from Livestock Feeding Operations";
- "Review of Emission Factors and Methodologies to Estimate Ammonia Emissions from Animal Waste Handling", April 2002 (EPA-600/R-02-0217); and
- On-going coordinated research by six universities, including the University of Minnesota on "Aerial Pollutant Emissions from Animal Confinement Buildings".

In addition to these studies, we intensively searched and reviewed new literature on ammonia emissions, including an analysis of the EPA National Emissions Trends estimates. Our literature review began with a search of bibliographic databases and the World Wide Web and resulted in the identification of eight significant, recently published works, listed in **Table 1-1**. Further, we contacted EPA and well-known researchers in the area of ammonia emissions for their recommendations, unpublished works, and works in progress. We obtained valuable information from the following individuals:

Urban Sources

- Joseph Somers, EPA, provided information for motor vehicles.
- Phil Lorang and Dennis Beauregard, EPA, provided information for POTWs.

Rural Sources

- David Allen, University of Texas at Austin; Steve Andersen, Texas Commission on Environmental Quality (TCEQ); and Alice Gilliland, National Oceanic and Atmospheric Administration (NOAA) provided information for livestock management. Dallas Safriet, EPA; Dr. David Terry, Association of American Plant Food Control Officials (AAPFCO); and Carl Wacker and Joe Mayfield, U.S. Department of Agriculture (USDA), provided information for fertilizer.
- Steve Andersen, TCEQ, provided information for soils.

There are several research efforts currently underway but are presently without preliminary results. The USDA and the EPA are working to refine the ammonia inventory for all source categories, including biogenic categories that are omitted from the EPA's most recent inventory. Recent research has shown that soil/plant canopy systems can act either as sources or sinks of ammonia (Roe and Mansell, 2001). Because of the sheer extent of land area, natural soil emissions could overwhelm ammonia inventories for some regions. For this reason, and due to a high degree of uncertainty in the estimated emissions from soils, soil emissions are often omitted from ammonia inventories. In addition, the California Air Resources Board (CARB) is currently evaluating several unquantified sources, such as biomass combustion (prescribed burning, wildfires, agricultural burning), composting, and residential wood combustion. Eventually, the results of these ongoing research efforts could be useful for further improvements to the CMU ammonia model.

Table 1-1. Recent research projects published from 1999-2002.

Topic	Title	Sponsor	Source
General	Nonpoint Source Ammonia Emissions in Texas: A First Estimate	TCEQ*	Corsi et al., 2002
General	Seasonal Ammonia Emission Estimates for the United States	NOAA, EPA	Gilliland et al., 2002
Livestock	Review of Emission Factors and Methodologies to Estimate Ammonia Emissions from Animal Waste Handling	EPA	U.S. Environmental Protection Agency, 2002a
Livestock	Emissions from Animal Feeding Operations	EPA	U.S. Environmental Protection Agency, 2001
Livestock	The Scientific Basis for Estimating Emissions from Animal Feeding Operations (Interim Report)	-	National Research Council, 2002
Livestock	Iowa Concentrated Animal Feeding Operations Air Quality Study	-	Iowa State University and The University of Iowa Study Group, 2002
Fertilizer	Joint EMEP/CORINAIR Atmospheric Emission Inventory Guidebook, Third Edition	EEA	European Environment Agency, 2001
Soils	Net Ammonia Emissions from Pine and Oak Forests in Texas	TCEQ*	Corsi et al., 2002

* Note: Until 2002, TCEQ was known as the Texas Natural Resource Conservation Commission (TNRCC).

1.3 GUIDE TO THIS REPORT

The remainder of this report is divided into sections covering the ammonia emissions from urban sources (Section 2) and rural sources (Section 3). An overall summary of findings is presented in Section 4 and additional information pertaining to calculation procedures for annualizing emission rates and details related to measurements of livestock emissions are provided in appendices.

2. URBAN SOURCES

Urban sources of ammonia include mobile sources, wastewater treatment plants, industrial point sources, and area sources. Mobile sources and wastewater treatment plants are usually the most significant urban sources and are present in all United States cities; therefore, they received the focus of our attention.

2.1 MOBILE SOURCES

Ammonia emission rates from internal combustion engines depend primarily on engine emissions control technology and secondarily on fuel type and other engine-design parameters. Catalytic converters, designed to reduce NO_x emissions from gasoline-powered vehicles, produce ammonia emissions as a by-product of NO_x conversion. As catalyst technology has advanced over the past twenty years, the conversion rate of NO_x to NH_3 has increased. Thus, late-model, gasoline-powered cars generate a larger fraction of ammonia emissions from on-road vehicle fleets.

2.1.1 On-road Motor Vehicles: Activity Data and Emission Factors

The CMU ammonia model is currently pre-loaded with existing county-level travel activity data for the entire United States. Future applications of the CMU tool will require that these data be updated accordingly for each vehicle type (e.g., gasoline-powered cars and trucks, diesel trucks). The Federal Highway Administration (FHWA) of the U.S. Department of Transportation provides state-level VMT data on its web site. County-level data are generally available from state departments of transportation (DOTs).

The CMU ammonia model employs emission factors of 0.0128 g/mile for cars and 0.068 g/mile for trucks. By contrast, the EPA currently applies an ammonia emission factor of 0.10 g/mile for three-way catalyst-equipped vehicles (U.S. Environmental Protection Agency, 2002). We found a range of emission rates during our literature review, which generally better matched the EPA's emission factor than that in the CMU model. Baum et al. (2001) determined a mean fleet ammonia mass emission rate of 0.15 g/mile (5% of NO_x emissions on a mass basis) at a Los Angeles freeway on-ramp; Fraser and Cass (1998) measured a mean emission rate of 0.115 g/mile for a fleet comprised of 91% catalyst-equipped vehicles; and Kean et al. (2000) measured an ammonia emission rate of 0.079 g/mile—or 10% of NO_x emissions on a mass basis—during a 1999 study in the Caldecott Tunnel (San Francisco Bay area). Only Gertler et al. (2002) measured ammonia emissions at much lower levels (0.015 g/mile) in the Tuscarora Tunnel in Pennsylvania. These seemingly inconsistent ammonia emission factor results may actually be the result of different driving conditions and different vehicle fleets. Shores et al. (2002) report that, under steady-state conditions, ammonia emissions are relatively small at 0.015 g/mile. Under hard acceleration, Shores et al. (2002) found ammonia emissions in the range of 0.23 to 0.33 g/mile.

Although the absolute emission rate of ammonia varied considerably from study to study, the ratio of NH_3-to-NO_x emissions had only a small variation (only between 5% and 10%).

Thus, we recommend updating the CMU model with a representative value of 7.5% to 8% of on-road gasoline-powered automobile NO_x emissions on a mass basis[1], which corresponds to the EPA's emission factor of 0.1 g/mile.

Additionally, Wilson et al. (2002) showed that ammonia emissions are inversely related to fuel sulfur content. Ammonia emission rates increase by as much as a factor of 7 with a corresponding factor-of-9 decrease in fuel sulfur content (from 324 ppm to 35 ppm). Future regulations are likely to impose increasingly strict limits on fuel sulfur contents. We produced a rough estimate of 1997 ammonia emissions from on-road motor vehicles by multiplying state vehicle miles traveled (VMT) estimates (U.S. Department of Transportation, 1997) with a conservatively high emission factor of 0.15 g/mile (Baum et al., 2001). **Figure 2-1** shows the results for the LADCO states (Illinois, Indiana, Michigan, Ohio, and Wisconsin) and surrounding states (Iowa, Minnesota, and Missouri), disaggregated into rural and urban components. These estimates could grow dramatically in the future—up to 75,000 metric tons per year for Illinois and Ohio, for example, or approximately equivalent to the magnitudes of livestock emissions in those states—due to the possibility that ammonia emissions from motor vehicles will increase when fuel sulfur contents decrease. *Thus, the impacts of new low-sulfur fuels should be considered in future-year estimates or forecasts of on-road motor vehicle ammonia emissions.*

2.1.2 On-road Motor Vehicles: Temporal Patterns

Activity patterns for on-road motor vehicles vary significantly by hour of day and day of week. In addition, ambient temperature affects motor vehicle NO_x emissions and may also impact ammonia emissions. Chinkin et al. (2002) analyzed diurnal, weekly, and seasonal patterns of traffic on southern California freeways and surface streets. Seasonally, traffic patterns varied little in southern California. However, this result may not be applicable in the Midwest, where people may have different habits because of differences in the climate and school-year calendar. *While the use of annual VMT data currently pre-loaded in the CMU model is probably sufficient, we encourage the use of seasonally specific local travel data available from most state DOTs.*

Figure 2-2 shows typical light-duty (LD) vehicle volumes by day of week and hour of day for freeways in the Los Angeles area of California. The Interior Basin and Long Beach sites represent urban activity patterns. At these locations, weekday LD vehicle volumes follow bimodal distributions with peaks during the morning and afternoon rush hours, while weekend LD vehicle volumes peak around midday. At the inflow/outflow sites (Indio and Castaic), weekend LD volumes follow an attenuated bimodal distribution. Volumes also are relatively high on Friday and Sunday afternoons. The increased volumes at inflow/outflow sites on Friday and Sunday afternoons are possibly a result of vehicles departing for and returning from recreational destinations outside the urban center. *We recommend applying these or similar temporal distributions to allocate weekly and diurnal emissions from mobile sources in the Midwest.*

[1] MOBILE6 reports the current gasoline fleet-wide average NO_x emissions of 1.19 gm/mile.

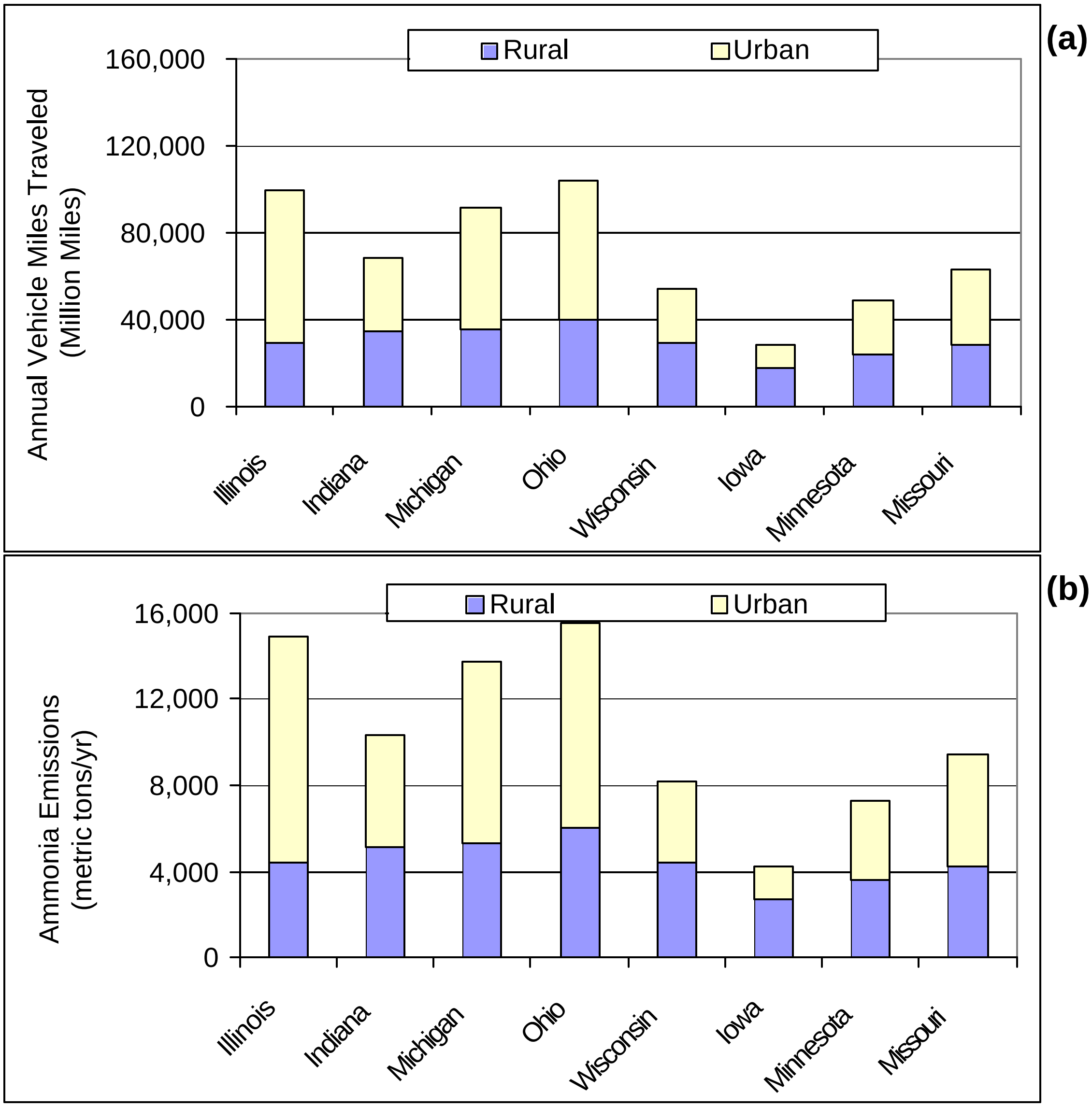

Figure 2-1. Annual 1997 VMT (a) and mobile source ammonia emissions (b) in metric tons per year (mt/yr) for the LADCO states (Illinois, Indiana, Michigan, Ohio, and Wisconsin) and surrounding states (Iowa, Minnesota, and Mississippi).

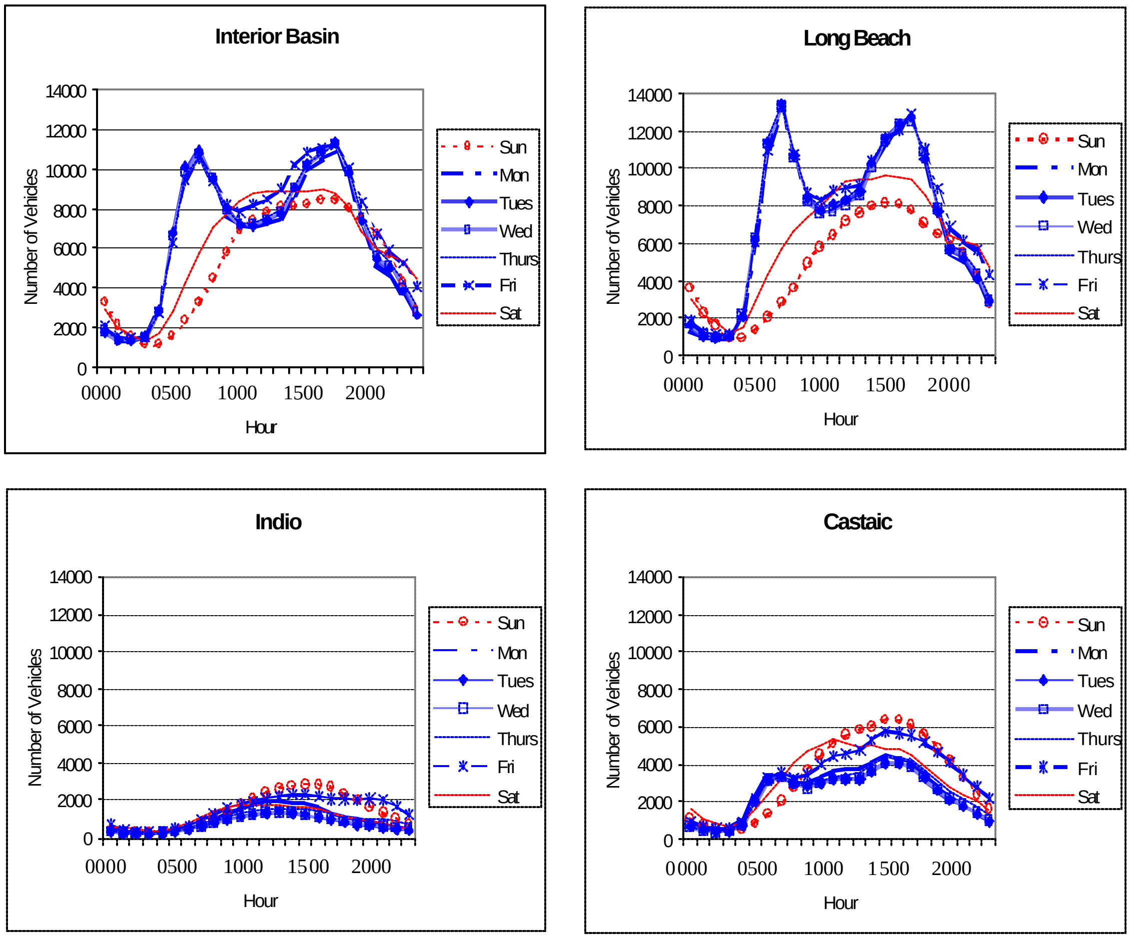

Figure 2-2. Average light-duty traffic volumes by hour of day and day of week observed at freeway sites in the near urban centers and outlying areas of Los Angeles, California.

2.1.3 Off-road Mobile Sources

The most recent research on ammonia emissions from off-road mobile sources that we identified was Harvey et al. (1983), who specified emission factors for off-road engines but with wide ranges of uncertainty:

- For gasoline-powered engines, 0.15 g NH_3/gallon
- For diesel-powered engines, 0.17g NH_3/gallon

These emission factors are currently in use by EPA for their internal inventory development efforts only. (At present, the EPA's NONROAD model does not produce emissions estimates for ammonia.) *Until improved emission factors or methods officially become available from EPA, we recommend augmenting the CMU model databases with the emission factors presented by Harvey et al. (1983) and with statewide estimates of off-road fuel consumption, which are reported annually by the Federal Highway Administration in its publication "Highway*

Statistics". Emissions may be disaggregated to the county level according to a spatial surrogate; as a simple solution, we suggest county-level NO_x emissions estimates that are produced by the EPA's NONROAD model.

2.2 WASTE DISPOSAL

2.2.1 Publicly Owned Treatment Works

Municipal sewage treatment plants process sufficient quantities of nitrogen-rich wastes to generate significant ammonia emissions under certain conditions. The EPA and the CMU ammonia model currently use an emission factor of 19 lb $NH_3/10^6$ gallons treated. This emission factor originated with the development of the 1985 National Acid Precipitation Assessment Program (NAPAP) Emissions Inventory (Warn et al., 1990). We evaluated this emission factor by contacting several EPA representatives and reviewing the original reference. Facing a lack of data, Warn et al. were forced to make a number of broad engineering assumptions in order to determine the emission factor. Warn et al. (1990) estimated their emission factor by applying the following elements of information:

- Survey data of 850 wastewater plants showed an average 75% difference between influent and effluent concentrations of ammonia-nitrogen at wastewater treatment plants (U.S. Environmental Protection Agency, 1985).
- A typical concentration (25 mg/L) of ammonia-nitrogen was assumed on the basis of information contained in a wastewater treatment plant engineering design manual (Metcalf and Eddy, Inc., 1979).
- At a pH of 7 to 8, ammonia air-stripping efficiency reaches only 10% (Lee and Naimie, 1985).

Warn et al. (1990) concluded that, at most, 18.75 mg/L of ammonia-nitrogen—the entire mass balance of influent-minus-effluent throughputs—is available for possible losses to the atmosphere through air stripping; and that because untreated municipal waste has a pH approximately equal to 7, 10% of this quantity will be lost in the aeration tanks that exist at most wastewater treatment plants. This equates to an emission factor of 1.875 mg/L (or in alternate units, 15.6 lb/MMgal or 7.10 kg/MMgal). This approach required three inherent assumptions, all of which we consider to be insupportable:

- We believe the loss of influent ammonia-nitrogen to the atmosphere is not a likely fate. By design, wastewater plants facilitate aerobic microbiological processes. Under aerobic conditions, ammonia-nitrogen is under heavy biological demand. The most likely fate of influent ammonia-nitrogen is uptake and consumption by microorganisms. Microbial uptake probably accounts for the vast majority of the observed differences between influent and effluent ammonia-nitrogen concentrations.
- We believe that air stripping, which occurs in aeration basins and/or trickling filters, is not necessarily the predominant source of ammonia emissions at wastewater treatment plants. Very little atmospheric ammonia losses should be expected from aerobic processes that are reasonably close to the typical design pH values of municipal

wastewater plants. However, anaerobic microbial processes are very likely to produce ammonia-nitrogen as a biological by-product of decomposition. If a wastewater plant has on-site anaerobic processes that are open to the atmosphere—and not all wastewater plants do—ammonia emissions should be overwhelming. In fact, we observed this to be the case at a small California municipal plant that maintained its own sludge-dewatering beds on site (Coe et al., 1998). The sludge beds were stagnant and rich in organic material; therefore, they attained strongly anaerobic conditions. By factors of 3 to 25, the highest ambient concentrations of ammonia at that plant were observed in the immediate vicinities of the anaerobic sludge drying beds.

- Air stripping efficiency of ammonia is dependent on the concentration of free aqueous ammonia ($NH_{3(aq)}$). The concentration of $NH_{3(aq)}$ will decrease by a factor of 10 with each 1-digit reduction in the pH so that equilibrium is maintained with aqueous hydrolyzed ammonia (NH_4^+). Over the pH range of untreated municipal waste (6.5 to 8), the concentration of $NH_{3(aq)}$ should be expected to vary by a factor of 30. Thus, the stripping efficiency also should be expected to vary widely from facility to facility.

For these reasons, we strongly recommend alternative emission factors for POTWs. For aeration basins, we recommend an emission factor that is consistent with measurement data collected by the Los Angeles County Sanitation District (LACSD) and the County Sanitation Districts of Orange County (CSDOC). The LACSD measured ammonia emission rates equivalent to approximately 50 to 150 lb per year from the aeration basins of two of its facilities (Knapp and Adams, 1997). These facilities process 30 to 60 million gallons per day. On the basis of these observations, emission rates for the aeration basins ranged from 0.002 to 0.01 lb/MMgal (or 0.001 to 0.006 kg/MMgal). These emission rates are about 3 orders of magnitude smaller than the emission factor that was formulated for the NAPAP inventory. The CSDOC measured emission rates at a variety of processes for two of their facilities, which processed from 75 to 175 million gallons per day (Kogan and Torres, 1997):

- Sludge dewatering—0.10 lb/MMgal (or 0.046 kg/MMgal)
- Air-activated sludge (aeration basin)—0.010 lb/MMgal (or 0.0045 kg/MMgal)
- Headworks—0.0097 lb/MMgal (or 0.0044 kg/MMgal)
- Primary clarifier—0.0013 lb/MMgal (or 0.00059 kg/MMgal)
- Oxygen-activated sludge—0.00010 lb/MMgal (or 0.000046 kg/MMgal)

From the CSDOC measurements, it is apparent that the sludge-dewatering process was the predominant source of ammonia in the processes that were studied. Emissions from the aeration basin and headworks were an order of magnitude smaller than those from the sludge dewatering. *Thus, we recommend an emission factor of 0.12 lb/MMgal (or 0.054 kg/MMgal)*. It should be noted that this emission factor may not be representative of all POTWs because the designs of sludge-dewatering and other operations vary from plant to plant. In addition, some plants capture and control offgases for some of their processes, such as anaerobic sludge digesters. Lastly, our recommended emission factor may not be comprehensive because it only covers the operations for which measurements were taken by the CSDOC. For example, some plants may employ additional sludge-handling processes, such as digestion, composting, and land application. Most likely, only individual facility operators will have access to the requisite information and data needed to estimate emissions from sludge-handling at their plants.

2.2.2 Landfills

The most recent research on ammonia emissions from landfills identified was Eggleston (1992), who suggests the ratio of ammonia to methane emissions from landfills is approximately *0.7%. We recommend augmenting the CMU model databases with this factor and with the EPA's annual estimates of methane emissions from landfills.* Emissions may be disaggregated to the county level with the same spatial distribution of methane emission inventory (by county or by the geographic coordinates of individual landfills.)

3. RURAL SOURCES

A wide variety of ammonia sources are found in rural areas, including livestock and poultry operations, fertilizer applications, and naturally occurring emissions from soils. Current ammonia inventories suggest that the most important rural categories are livestock and poultry operations and fertilizer applications. Livestock and poultry operation and fertilizer application are estimated to comprise about 85% of total national ammonia emissions in the United States in 1998 (U.S. Environmental Protection Agency, 2002b).

3.1 LIVESTOCK AND POULTRY

Livestock and poultry emissions are estimated to be the most significant sources of ammonia emissions nationwide (U.S. Environmental Protection Agency, 2002b). Nationally, the EPA estimates that ammonia emissions from commercial animal husbandry in the United States is dominated by calves and cattle (78%), followed by pigs and hogs (19%). The other 3% of emissions arise from chickens (2%) and sheep (1%). Livestock and poultry populations vary greatly from state to state for a variety of reasons such as climate and soil characteristics; thus, locally derived livestock headcounts are needed.

To better understand the magnitude of livestock and poultry emission categories, we estimated statewide ammonia emissions for each livestock category using head count information at the state level for 1997 (Oregon State University, 1999) and EPA default ammonia emission factors (Battye et al., 1994). **Figure 3-1** shows estimates of ammonia emission from calves and cattle, pigs and hogs, sheep, and chicken/pullets for the LADCO states (Illinois, Indiana, Michigan, Ohio, Wisconsin) and surrounding states (Iowa, Minnesota and Missouri). Of these Midwestern states, Iowa has the largest amount of animal husbandry emissions at 224,000 metric tons per year (almost twice that of any other Midwestern state). Michigan has the smallest amount of animal husbandry emissions at 34,000 metric tons per year (about one-half that of any other Midwestern state). Wisconsin is the only Midwest state with a higher contribution from cattle and calf emissions (91%) than the national average (78%). Iowa (60%), Indiana (58%), and Illinois (56%) have hog and pig emission contributions 2.5 to 3 times the national average (19%). Sheep represent 1% or less of total animal emissions within each Midwest state, which is similar to the estimated sheep emission contribution for the entire United States. Emissions from poultry are 2% or less in each state, similar to the national average, with the exception of Ohio (6%), Michigan (3%), and Indiana (6%).

3.1.1 Livestock and Poultry Emission Factors

A wide variation in livestock emissions is reported in different studies in the United States (U.S. Environmental Agency, 2002a; U.S. Environmental Agency, 2001; Corsi et al., 2000) and Europe (European Environment Agency, 2001; Sutton et al., 1995; Schmidt, 1996; Battye et al., 1994; Asman, 1992; Buijsman, 1987). Recommendations for the use of the CMU emission factors and the EPA livestock emission factors (U.S. Environmental Protection Agency, 2002b) are derived from the 1994 Battye et al. report. These emission factors are shown in boldface type under the Battye et al. column in **Table 3-1**. The 1994 Battye et al. report recommended that European animal waste ammonia emission factors developed by Asman (1992) be used in the United States. Asman defined 21 animal categories and sub-categories and three broad waste management categories (stable and storage; spreading; and grazing). The emission factors in Asman's paper were based on tests that were conducted in the Netherlands in the late 1980s by various researchers and were developed by dividing the emission of a category by the number of animals in that category. The main limitations of the Asman emission factors are that animal weight and climate factors (expressed in seasonal and diurnal temperature variation) are not taken into account.

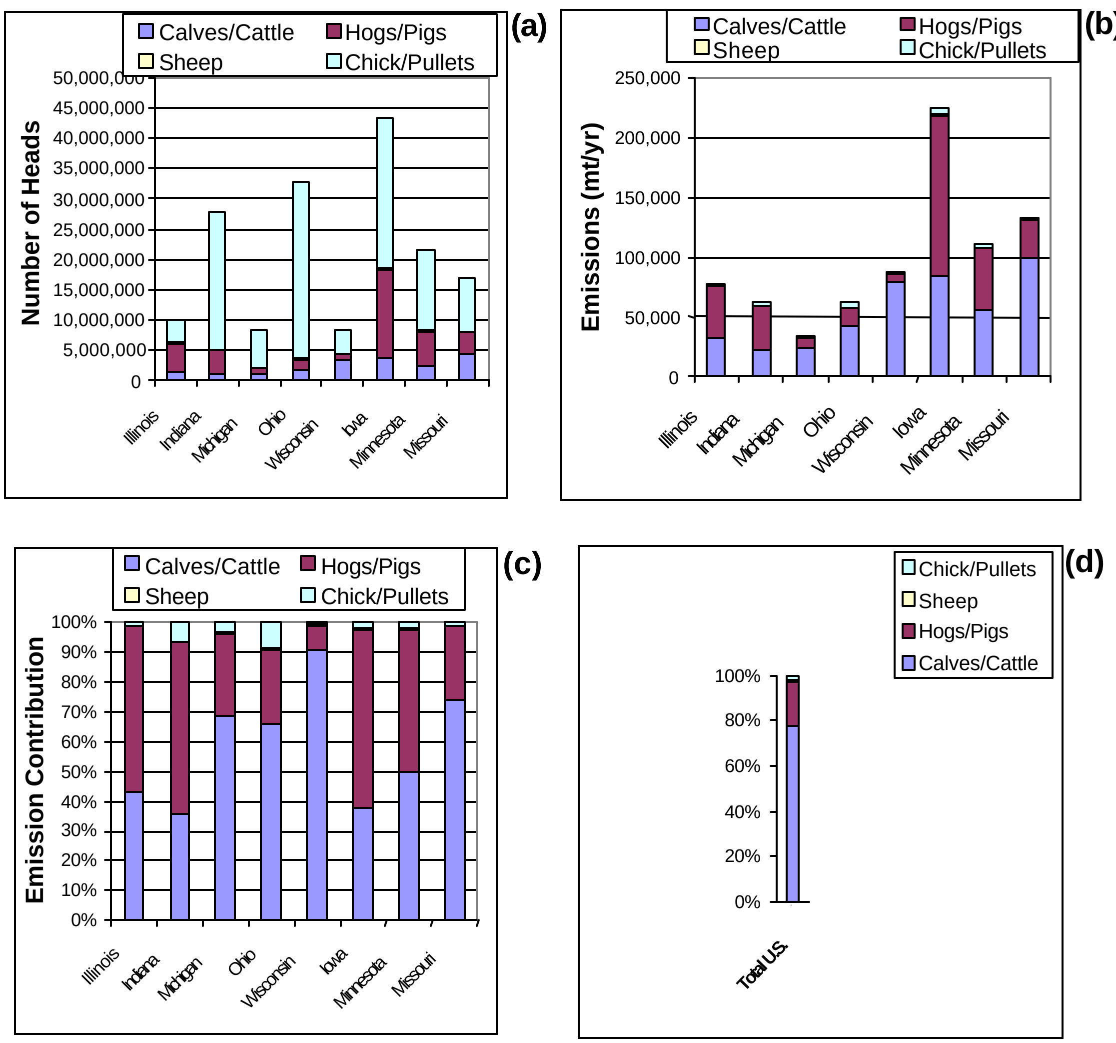

Figure 3-1. Livestock (a) population and (b, c) ammonia emissions statistics for the LADCO and surrounding states and (d) ammonia emissions contributions by animal type for the United States.

Table 3-1. Alternative emission factors (lb/head-yr) for livestock.

Animal	Battye et al. 1994[a]	Sutton et al. 1995	Corsi et al. (2000)	U.S. EPA (2001)	U.S. EPA (2002a)	EEA (2001)
Cattle – Dairy		48-72	72 (43-101)	51	55	24.6
Beef			34 (18-103)	45	20	12.3
Heifers						12.3
Steers						12.3
Cattle and Calves – Composite	50.5	37 (17 - 54)				
Cows and heifers that have calved (beef cows)	**87.57**					
Cows and heifers that have calved (milk cows)	**87.57**					
500 lb. and over: Heifers - beef cow replacements	33.49					
500 lb. and over: Heifers - milk cow replacements	**28.75**					
500 lb. and over: Heifers – other	28.75					
500 lb. and over: Steers	**18.12**					
500 lb. and over: Bulls	61.53					
Calves under 500 lb.	11.53					
Horses	**26.9**	22 (11-44)				5.1
Sheep	**7.43**	2.4 (0.88-3.3)				0.46
Goats						0.46
Hogs	**20.30**	9.5 (6.8-12.5)	12 (2.4-24)	5.8	15 ± 4.4	6.39/16.43
Sows farrowing	35.56					
Other – breeding	11.5					
Market hogs - under 60 lb.	15.4					
60-119 lb.	15.4					
120-179 lb.	24.3					
180 lb. And over	24.3					
Chickens – composite	**0.393**	0.48 (0.33-0.66)			0.22	
Chickens – broiler	0.368		0.51 (.31-.71)	0.5	0.22	0.28
Laying	1.32		0.97 (.37-1.6)	0.7		0.37

Table 3-1. Alternative emission factors (lb/head-yr) for livestock.

Animal	Battye et al. 1994[a]	Sutton et al. 1995	Corsi et al. (2000)	U.S. EPA (2001)	U.S. EPA (2002a)	EEA (2001
Pullets			0.37			
Pullets - laying age	.672				0.22	
- over 3 mos., not laying	.593					
- under 3 mos.	.375					
Other chickens	.395					
Turkeys	1.89	1.5 (1.1-2.1)	1.9 (1.2-2.6)	2.0		0.92
Young turkeys	1.96					
Old turkeys	2.82					
Fryer-roaster	1.89					
Geese						0.92
Ducks		0.22 (0.13-0.29)	0.26 (.15-.35)			0.92

[a] Battye et al. Recommended emission factors developed by Asman (1992).
Note: Ranges represent high-low recommended emission factors collected from several sources.

A review of international scientific literature published since 1994 revealed a few European papers that included new field test data on ammonia emissions. In addition, several summary papers were found that provide reviews of emission factors or emission estimates, and ammonia emission estimates using existing emission factors. These and other papers and the citations therein indicate that emission factor research is ongoing, especially in the Netherlands and, to a lesser degree, in the United Kingdom and Denmark.

The Dutch Institute for Health and Environment (RIVM) developed a comprehensive methodology to estimate ammonia emissions from animal manure, fertilizer usage, industrial processes, and households. This methodology was published in 1994. In 1998, the ammonia emission methodology underwent a comprehensive review by researchers associated with the Ministry of Agriculture and Fisheries because the level and trend of estimated emissions from agricultural sources were consistently and significantly different from emissions back-calculated from ambient air data. The comprehensive review included the most recent literature and expert knowledge available in the Netherlands at that time.

The limited work in Denmark led the Danish Government to follow a mass balance approach. The Danish approach was developed to make best use of the available activity data in Denmark. Ammonia emissions were calculated separately for housing and manure storage and during and after spreading. Animals were divided into 31 categories according to species and housing type. The categories were chosen to match available national activity data from the Danish Agricultural Advisory Centre. The total Danish livestock and poultry population was distributed among these categories, and the total nitrogen excreted annually by the animals in each category was calculated by multiplying the animal numbers by the annual nitrogen excretion per animal. The fate of this nitrogen was followed throughout the manure-handling chain, with ammonia emission calculated as a percentage of the amount of nitrogen present in each link in the chain. The model takes into account additional nitrogen from bedding and spilled feed. Emission factors are expressed as the percent of total remaining ammonia and nitrogen.

Before applying European emission factors to the United States, it is important to acknowledge that there are differences in both animal waste management practices and animal husbandry practices in Europe and the United States. For example, animal waste in the United States is commonly stored in lagoons which are uncommon in Europe; in Europe, waste is more commonly stored in concrete tanks. Following are two examples of animal husbandry that differ between Europe and the United States:

- Ranches in the United States are generally larger in size and enable wider cattle grazing activity. Because confined cattle, fed food high in nitrogen as is more common in Europe, emit more ammonia, the use of European emissions factors may be overstating cattle emissions in the United States.

- Pasture emissions are affected by the weather (ambient temperature, precipitation) and soil conditions. Weather and soil conditions corresponding to measurements made in Europe may be substantially different from those in the Midwest.

These practices could affect the applicability of emission factors based on European practices.

Given these differences and others, it may be concluded that the Asman (1992) emission factors recommended by Battye et al. (1994) may not be well-suited for estimating emissions in the United States. Yet, during the late 1990s, they continued to be used in the United States for lack of better data (U.S. Environmental Protection Agency, 2002a).

Overall, the cattle, swine, and poultry emission factors recently reported by EPA's Office of Research and Development (ORD) (U.S. Environmental Protection Agency, 2002a) and the European Environment Agency (2001) are reasonably equivalent. However, the emission factors cited by ORD are different than those used by the CMU model and those cited elsewhere by EPA (U.S. Environmental Protection Agency, 2002b). CMU is currently using a dairy cow emission factor of 87 lb/head/yr compared to ORD's cited 50 lb/head/yr factor. The beef cattle emission factor cited by ORD is 20 lb/head/yr or about 65% lower than the 87 lb/head/yr factor in the CMU model. Additionally, the EPA recently recommended using a 25% lower pig and hog emission factor of 15.4 lb/head/yr based on a measurement study in North Carolina (U.S. Environmental Protection Agency, 2002a). The prior EPA pig and hog ammonia emission factor of 20.3 lb/head/yr was based on the 1994 European report (Battye et al., 1994).

In European studies (European Environment Agency, 2001), dairy cows were found to excrete on average 220 pounds of nitrogen per year, about 2.25 times more than nitrogen excreted from beef cows. Thus, assuming that ammonia emissions are proportional to excreted nitrogen, ammonia emissions from dairy cows should be about 2.25 times greater than from beef cattle. The recent ORD (U.S. Environmental Protection Agency, 2002a) and Corsi et al. (2000) emission factors for dairy cows and cattle are consistent with this difference of about a factor of 2.25. We prefer the ORD emission factors for several reasons: they are consistent with those reported by the European Environment Agency (2001); ORD relied on a detailed mass balance explanation for arriving at the emission factors; and ammonia emissions represent about 25% of nitrogen excreted, roughly in line with present thinking.

Therefore, we recommend emission factors for cattle, swine, and poultry consistent with those determined by ORD (U.S. Environmental Protection Agency, 2002a). To estimate the potential effect of these changes, we applied simple scaling factors to the existing CMU emission estimates and found that the net effect is a reduction of cattle and swine emissions by 25% to 65%. Note that, independently, Gilliland et al. (2002) using inverse modeling found that a similar reduction in cattle and swine emission factors is needed to properly account for observed concentrations. They found that the overall 1990 annual ammonia National Emission Inventory (NEI) should be approximately 35% lower minimize errors between modeled and observed wet [NH4+]. A 35% overall emission decrease nationwide can be obtained by applying the ORD (U.S. Environmental Protection Agency, 2002) reduced cattle (dairy and beef) and swine emission factors relative to that used in the NEI (Battye et al., 1994).

Because the ORD report does not include updated emission factors for horses, sheep, and goats, *we recommend emission factors reported by the European Environment Agency (2001) for these animals*. Emission factors for these categories are highly uncertain as depicted in Table 3-1 (U.S. Environmental Protection Agency, 2001).

Animal emission factors in general are not well-characterized. The National Academy of Science (NAS) (2002) reported that ammonia emission factors increase as animals age, differ due to the manure storage system, and are affected by climate, including temperature and moisture. Because these issues are not systematically being taken into account and perhaps because a wide range of emission factors were encountered in the literature, the NAS concluded that it is unreasonable to expect any single emission factor determined over a short period of time will represent a reliable annual emission factor (National Academy of Science, 2002).

Thus, although we recommended the use of the latest EPA swine emission factor (U.S. Environmental Protection Agency, 2002a), the representativeness of this emission factor is questionable because it represents a single point-in-time and space measurement. It should not be considered equivalent to an annual emission factor. Nevertheless, because of a lack of other data, the EPA recommends it for application. Indeed, all alternative emission factors (see Table 3-1) are equal to a single measurement or a composite of measurements primarily made in the summer. We expect from scientific principles that ammonia emissions increase with ambient temperatures and greater wind speed. Therefore, the use of emission factors that are primarily based on summertime measurements are likely to produce an overestimate of total annual emissions. Appendix A describes a scientific approach to obtain an annual emission factor from a local point-in-time measurement.

3.1.2 Livestock and Poultry Activity Data

No single source of livestock and poultry activity data is currently and consistently available. However, *the combination of U.S. Agricultural Census Data (which is renewed every five years) and, during the intervening periods, National Agricultural Statistics Service (NASS) estimates (which are generated annually) is recommended for use*. Note that the CARB identified limitations of the U.S. Agricultural Census and NASS data because they do not reflect some factors that affect actual state cattle populations, such as seasonal import and export of animals to other states (Gaffney and Yu, 2002). The CARB has been developing methods to adjust estimates derived from the U.S. Agricultural Census and NASS data so that they better reflect real-world populations (Gaffney and Yu, 2002). The CARB's adjustment factors significantly affect ammonia and methane emissions estimates for California, and it is likely that this may also be a significant issue in the LADCO states. However, there is insufficient information to propose a generally applicable adjustment method to the available information at this time.

USDA statistical information is cited by Pierce and Bender (1999) for estimating the annual livestock population. The Government Information Sharing Project (Oregon State University, 1999) maintains records of animal headcount data for 1987, 1992, and 1997.[2]

3.1.3 Temporal Variations

Seasonal Allocation

As noted above, no seasonal emission factor data are available. *Therefore, we recommend the seasonal distribution in emissions proposed by Gilliland et al. (2002)* which is based on modeled results. While Gilliland et al. (2002) present quantitative results, they recommend emphasis be placed on qualitative conclusions, such as seasonal variations in emissions. They recommend emphasis be placed on qualitative rather than quantitative findings because substantial biases and uncertainties exist in their approach. The uncertainties within their air quality model can influence the results, and not all model uncertainties can be included when estimating *a posteriori* ammonia emissions. Furthermore, these results are specific to the domain and time period considered. We acknowledge that there are concerns in the use of modeled outputs to adjust emissions rates and that the development of improved methods should be a high priority. However, seasonal measurements conflict (as reported in Appendix B), and a non-varying seasonal distribution is even less likely to reflect real-world conditions than is Gilliland's inverse-modeled distribution. Because livestock is such a predominant source of emissions, we feel that it is critical to develop the most plausible representation of real-world conditions, in spite of the known limitations.

Use of the Gilliland et al. (2002) monthly/seasonal scaling factors results in an emission decrease in the late spring and an emission increase during summer relative to annual average emissions. Since the Gilliland et al. (2002) factors were derived prior to the newer and lower ORD-recommended emission factors (U.S. Environmental Protection Agency, 2002a), they need

[2] In some states, county activity data are not reported or were withheld, but the state total is reported. When this occurs, the difference between the state and those counties with activity data must be determined. This difference is apportioned to each county without activity data. There are cases in which activity data are reported under a general county code designation of all other counties. Data reported under this county code may be added to the withheld totals for the state before distributing the state totals to counties. There are several states that withhold state-level activity data. In these cases, State totals are first estimated by calculating the total activity corresponding to all states combined that withhold data. This value is calculated by subtracting the category-specific totals from all states that reported data from the national total. The remaining activity data are then distributed equally or relative to size or in some other manner, and then distributed to each county.

to be corrected for the current emission factor recommendations. Accounting for the current ORD emission factors (U.S. Environmental Protection Agency, 2002a), the Gilliland et al. (2002) inverse factors were corrected accordingly by a factor of 1.5. The resulting smoothed seasonal adjustment factors are shown in **Table 3-2.**

Table 3-2. Qualitative monthly scaling factors to apply to 1990 Annual Ammonia Emission Inventory (Gilliland et al., 2002).

Month	Recommended Seasonal Allocation Factors (x Annual Average Emission Rates)
January	67%
February	75%
March	75%
April	82%
May	126%
June	164%
July	183%
August	154%
September	115%
October	73%
November	51%
December	51%

Another source of seasonal variations in emissions is migration. The CARB reports that cattle are brought to California during the winter from other states for grazing purposes (California Air Resources Board, 1999). These livestock may be temporally imported from midwestern states before the cold weather arrives and returned in the spring. This additional seasonal factor should be further investigated in the future.

Diurnal Allocation

Based on studies by Aarnink (1997) and Harris (2001), who both reported that emissions from pig houses varied diurnally, *we are recommending that a diurnal profile be applied for swine emissions*. Aarnink (1997) reported that ammonia emissions from houses with rearing pigs and houses with fattening pigs had higher emissions during the day than during the night: +10% for rearing pigs and +7% for fattening pigs. For rearing pigs, emissions peaked in the morning, but for fattening pigs they peaked in the afternoon. This information was used to develop the interim empirical diurnal profile for ammonia emissions from pigs shown in Table 1-4.

Unfortunately, (as illustrated in Appendix B), consistent information is not available to determine diurnal profile of emissions from dairy cows and cattle or other livestock. For this reason and because a diurnal profile is expected from other livestock categories, *STI recommends use of the swine diurnal profile for all livestock categories in the interim* until better information becomes available for these other categories.

3.2 FERTILIZER

Fertilizer emissions are the second most abundant source of ammonia emissions nationwide, (U.S. Environmental Protection Agency, 2002b). Few early studies (pre-1996) provided details of the basis for their recommended annual emission factors, with the exception of Battye et al. (1994). Moreover, the European Environment Agency (2001) now considers the earlier work of Buijsman et al. (1987) to be out of date and believe that the earlier reported emission factors overestimate ammonia emissions from fertilizer applications.

Historically, the EPA recommended the use of the Battye et al. (1994) emission factors, in part, because they were accompanied with supporting data and an explanation of factor development. These emission factors range from 24 lb to 364 lb of NH_3 emitted per ton of nitrogen fertilizer applied (or equivalently, 1% to 15% of NH_3 as nitrogen emitted per ton of nitrogen fertilizer applied). The emission factors in the CMU model are similar to those recommended by the EPA and reported by Battye et al. (1994) as listed in **Table 3-3**. Of the twelve fertilizer emission factors used in the CMU tool, the highest ammonia emission factor of 15% is assigned to urea fertilizer and among the lowest is the value of 2% assigned to ammonium nitrate. (Note: the CMU model documentation incorrectly identifies these fertilizer emission factors as ranging from 0.01% to 0.15%).

Corsi et al. (2000) recommended emission factors similar to those reported by Battye et al. (1994) except for an emission factor 4 times greater from anhydrous ammonia, an emission factor equal to two-thirds that of urea, and an emission factor 1.4 times that of ammonia sulfate (see Table 3-3). Subsequently, Corsi et al. (2002) reported that in alkaline agricultural soils ($pH > 7$), a significant fraction of the ammonium and urea fertilizers applied are lost to the atmosphere (e.g., typical average of 20% of the amount applied) although losses from forest fertilization are often lower.

In general, ammonia emissions from fertilizer are a function of the fertilizer type; application type (injection or surface); application rate (e.g., 100 kg nitrogen/hectare [N/ha] or 250 kg N/ha); soil type; and climate. We could identify only one reference source (European Environment Agency, 2001) that accounts for differences associated with fertilizer type, soil type, and climate. *STI recommends use of the European Environment Agency (2001) emission factors because they are fertilizer type-, soil type-, and climate-dependent.* The basis for fertilizer emissions varying with soil and climate is well-established (e.g., crop-related emissions are greater in warmer climates and soil emissions (direct fertilizer losses) generally increase at higher soil pH. However, since generalized rates are required, the European Environment Agency developed the following classification system:

- Group I - Warm temperate areas with a large proportion of calcareous soils (e.g., Greece, Spain). Subtropical and continental climates would be expected to fall into this Group.
- Group II - Temperate and warm-temperate areas with some calcareous soils (or managed with soil pH >7), but with large areas of acidic soils (e.g., Italy, France, United Kingdom, Ireland, Portugal, Belgium, Netherlands, Luxembourg).
- Group III - Temperate and cool-temperate areas with largely acidic soils (e.g. Scandinavian countries, Germany, Switzerland, Austria).

Table 3-3. Alternative emission factor (not necessarily annual emission factor) recommendations for fertilizer application.

Fertilizer Type	Battye et al., 1994 kg NH_3/Mg N	Weerden and Jarvis , 1997	CMU Emission Factor	Corsi et al., 2000 kg NH_3/Mg N	EEA, 2001 Group I % of total N applied	EEA, 2001 Group II % of total N applied	EEA, 2001 Group III % of total N applied
Anhydrous Ammonia	12 (1%)		***1%***[a]	49 (12-121)	4%	4%	4%[b]
Aqua ammonia	12 (1%)		***1%***[a]	12			4%[c]
Nitrogen solutions	30 (2.5%)		8%[a]	30	8%	8%	8%[b]
Urea	182 (15%)	12-23%	15%[a]	121 (61-279)	20%	15%	15%[b]
Ammonium nitrate	25 (2.1%)	1-1.6%	***2%***[a]	24 (10-121)	3%	2%	1%[b]
Ammonium sulfate	97 (8%)		***8%***[a]	140 (97-182)	15%	10%	5%[b]
Calcium ammonium nitrate			***2%***[a]	24	3%	2%	1%[b]
Ammonium thiosulfate	30 (2.5%)		2.5%[a]				2.5%[a]
Other straight nitrogen	30 (2.5%)			30			
Ammonium phosphates	48 (4%)		***4%***[a]	55 (49-61)	5%	5%	5%[b]
N-P-K[a]	48 (4.8%)			49	3%	2%	1%[b]
Potassium nitrate			***2%***[a]				1%[b]
Miscellaneous (spring)			***4%***				7%[d]
Miscellaneous (fall)			***15%***				7%[d]

Note: % represent the percent loss of the nitrogen-content that the corresponding emission factor represents.

[a] Battye et al. (1994).
[b] European Environment Agency (2001).
[c] Equal to anhydrous ammonia.
[d] Weighted average.

CMU emission factors that STI recommends be changed

Our literature review revealed a considerable range in reported emissions rates. However, Weerden and Jarvis (1997) reported loss rates of 12% and 23% for ammonia from arable and grassland soils with urea, respectively, in the United States, and rates of 1% and 1.6% of the nitrogen amount applied and ammonia emission factors from ammonium nitrate applied to arable and grassland soils, respectively. These emission factors are consistent with the range of Group I to Group III factors recommended by the European Environment Agency (2001). Thus, based on limited evidence, the European factors appear appropriate for use in the United States.

The principle requirement to use the European Environment Agency (2001) system in the LADCO states is to select those most representative of the LADCO area. To fully implement the European approach ultimately requires the use of an integrated geographic information system (GIS). The USDA GIS databases could be used to identify cropland areas (1) with calcareous soils and (2) in warm-temperate, temperate, or cool-temperate climates. STI realizes that implementing a full GIS approach will take more time than is available to meet the immediate needs of LADCO for an emissions estimation tool. Therefore, our proposed recommendations are suitable for the interim period until a GIS approach is feasible. As illustrated in **Figure 3-2**, the LADCO states appear generally best characterized as cool-temperate areas. For this reason, STI recommends the use of the European Environment Agency Group III fertilizer emission factors that are recommended for cool-temperate areas (European Environment Agency, 2001) to estimate ammonia emissions from fertilizer application throughout the LADCO states.

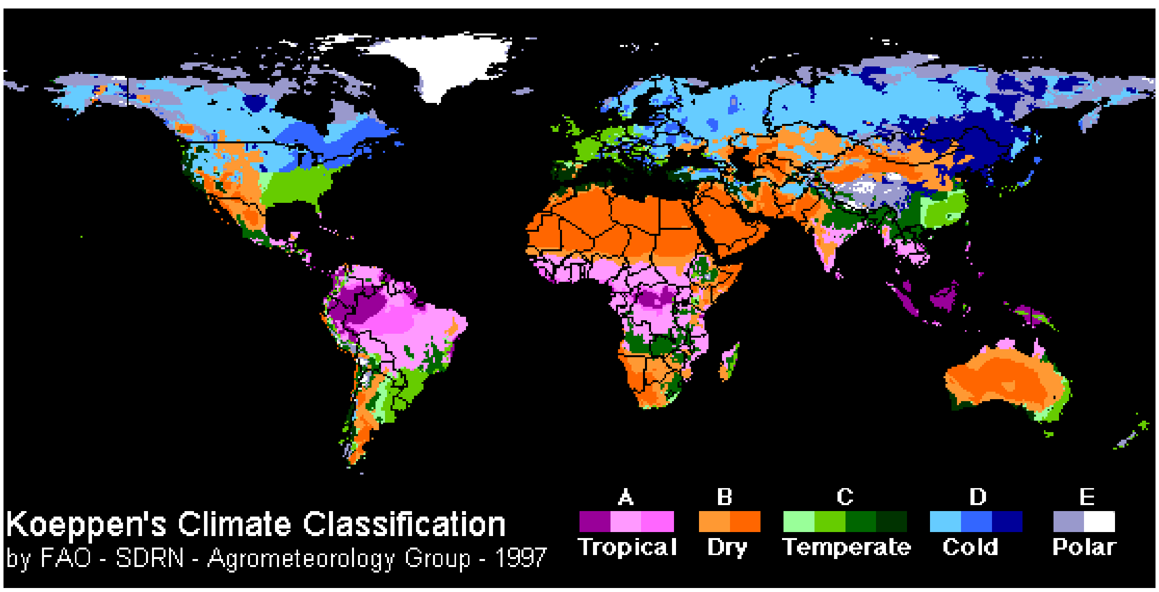

Figure 3-2. Koeppen's climate classification.

Additionally since the European Environment Agency (2001) factors represent annual emission factors, we recommend them over alternative emission factors appearing in Table 3-3 because they are limited to local point-in-time measurements. The adjustments to annualize point-in-time emission factors are shown in Appendix A. It appears from our literature review that most studies do not account for this adjustment to use point-in-time measurements to represent annual emission factors. STI is concerned that many of the reported emission factors appearing in Table 3-3 reflect measurements made over brief periods of time, often less than seven days. Measurements made during brief time periods and assumed to represent total emissions for the year may severely misstate annual emissions.

3.2.1 Fertilizer Activity Data

National fertilizer use data are available from the Association of American Plant Food Control Officials (2002). These data contain county-level usage of over 100 different types of fertilizers, including those that emit ammonia. Additional information is available by crop type from The Fertilizer Institute (2002). It can be used along with crop statistics from the U.S. Department of Agriculture (2002) to estimate the relative amounts of commercial fertilizer application by county.

We recommend reliance on crop-specific crop calendars, crop-specific fertilizer-use intensities, and county-specific crop acreage as the basis of temporal distributions, rather than use of semi-annual sales distributions, to partially reflect the seasonal distribution of use. We believe that this should reconcile potential issues related to time-of-use versus time-of-sale. This will involve acquiring recent fertilizer usage data and reprocessing the data to develop annual consumption figures, possibly as time-series averages (from 3 to 5 years). Total consumption for each county can be disaggregated proportionally to each crop grown according to the intensities of fertilizer use by each crop and the total acreage of these crops. Then, the crop-associated fertilizer consumption can be temporalized and re-aggregated to the county level in order to generate the temporal distribution for the county as a whole.

3.2.2 Temporal Variations

Because fertilizer use is seasonal and fertilizer-related ammonia emissions primarily occur during the short period of time following application, the fertilizer category could be an important source of emissions during the growing season. The CMU ammonia model incorporates the use of crop timing (planting) data to develop a monthly profile of ammonia emissions from fertilizer applications. Strader et al. (2002b) report two peak seasons of ammonia emissions from fertilizer. In the Midwest, especially in the northern areas, the spring peak begins in April with a comparable peak in the fall.

For farms in Minnesota, Bruening (2000) reports 49% of anhydrous ammonia fertilizer was applied in fall, 36% was applied in spring, and 15% was side-dressed during the growing season. This alternative application rate should be compared to rates currently used in the CMU ammonia model. If there are significant differences between the Bruening rates and those used in the CMU ammonia model, further study of this issue is warranted. In the absence of local data, Pierce and Bender (1999) proposed a fertilizer seasonal allocation scheme equal to 10% in winter, 50% in spring, 30% in summer, and 10% in fall.

Midwest Research Institute (1998) found that hourly emission rates of ammonia from fertilizer applications exhibit diurnal patterns that follow temperature patterns. Anderson and Levine (1987) found a similar pattern in diurnal nitric oxide fluxes from soil. Because of its quantified nature, the diurnal nitric oxide emissions data were used to create the *diurnal profile* *recommended for use for ammonia and the data are shown in **Figure 3-3***. It is acknowledged that ammonia emissions data are a preferred source relative to nitric oxide emissions data, but none was found.

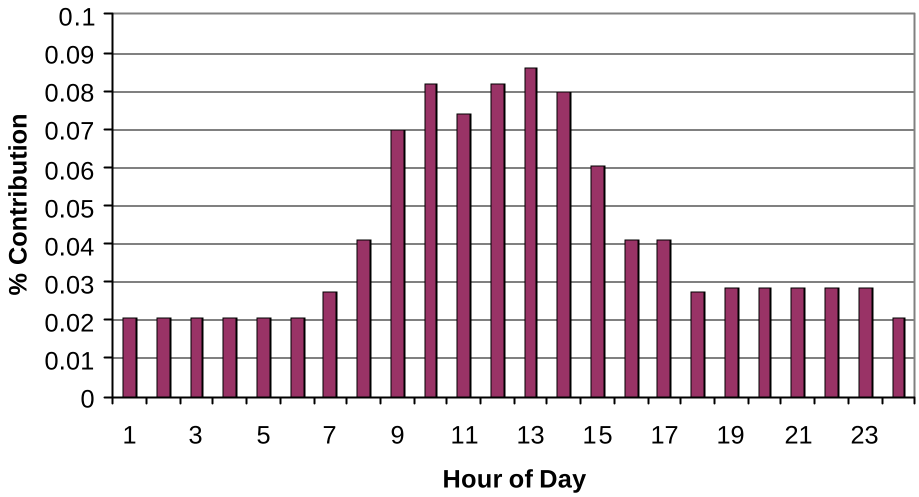

Figure 3-3. Diurnal ammonia emissions profile recommended for interim use.

First Principle Models

Potter et al. (2001) developed a statewide inventory of ammonia emissions from native soils and crop fertilizers in California using a first principle model. Because documentation was unavailable on the model itself, STI is unable to identify the scientific quality of the model at present, but further investigation of this model is suggested.

Alternatively, Cohen and Ryan (1990) developed a soil model which is the only diurnally varying emissions model of which we are aware that incorporates first principle concepts and accounts for emission changes resulting from diurnal temperature and moisture changes with soil depth and with changes in ambient weather conditions. In the absence of information about the Potter et al. (2001) model approach, we recommend use of the Cohen and Ryan (1990) model as a better longer-term solution for estimating the diurnal ammonia emission profile expected from fertilizer applications. Cohen and Ryan (1990) modeled ammonia transport within the soil using Equation 3-1:

$$\partial C_a / \partial t = D(T, \varepsilon, \theta_a, \theta_w, v_w) dC^2 / dz^2 - v_w H_{wa} dC_a / dz + \text{Production} - \text{Loss} \qquad \textbf{(3-1)}$$

where:

$$D = \text{soil diffusion coefficient} = \frac{(\theta_a D_a(T)/\tau_a + H_{wa}(T)\theta_w D_w(T)/\tau_w + 1 v_w H_{wa}}{(\theta_a + H_{wa}(T)\theta_w + H_{sw}(T) H_{wa}(T)(1 - \theta_w - \theta_a)}$$

in which θ_a and θ_w are the air and water content of the soil, which are time and depth dependent. $D_a(T)$ and $D_w(T)$ are ammonia molecular diffusion coefficients in air and water at temperature T. Molecular diffusion transport is slowed by the packing of the soil and is a function of the percentage of the soil occupied by the air and water (τ_a, τ_w). The chemical equilibrium concentration of ammonia between in air, water, and solid phases must also be determined ($H_{wa}(T)$, $H_{sw}(T)$). In addition, soil-water movement (l v_w) can also be an important mechanism of ammonia volatilization.

Based on knowledge of the soil, air, and water content and temperature with depth and time, emissions may be estimated using Equation 3-2:

$$\text{Emissions } (g/m^2/s) = k_a(m/s)\ (C_{sa}(g/m^3) - C_a(g/m^3)) - \text{Vegetation Uptake} \qquad \textbf{(3-2)}$$

where:

k_a = Air-side mass transfer coefficient, f(U,T)
C_{sa} = Ammonia concentration in soil-air at soil surface
C_a = Ammonia concentration in atmosphere 10 m above soil surface

To effectively utilize this approach in the LADCO area, hourly (diurnal) measurements are needed along with surface meteorological measurements for a year for at least one location; the recommendation is to obtain such information for a variety of locations and soil types. The soil types recommended would be selected in consultation with USDA personnel. **Figure 3-4** illustrates the variety of major soil types found throughout the world to gain some perspective on the need to gather data for at least each of the major soil types found in the LADCO states, recognizing, of course, that this is a simplification of actual emission variations over all the LADCO states due to climate/weather differences.

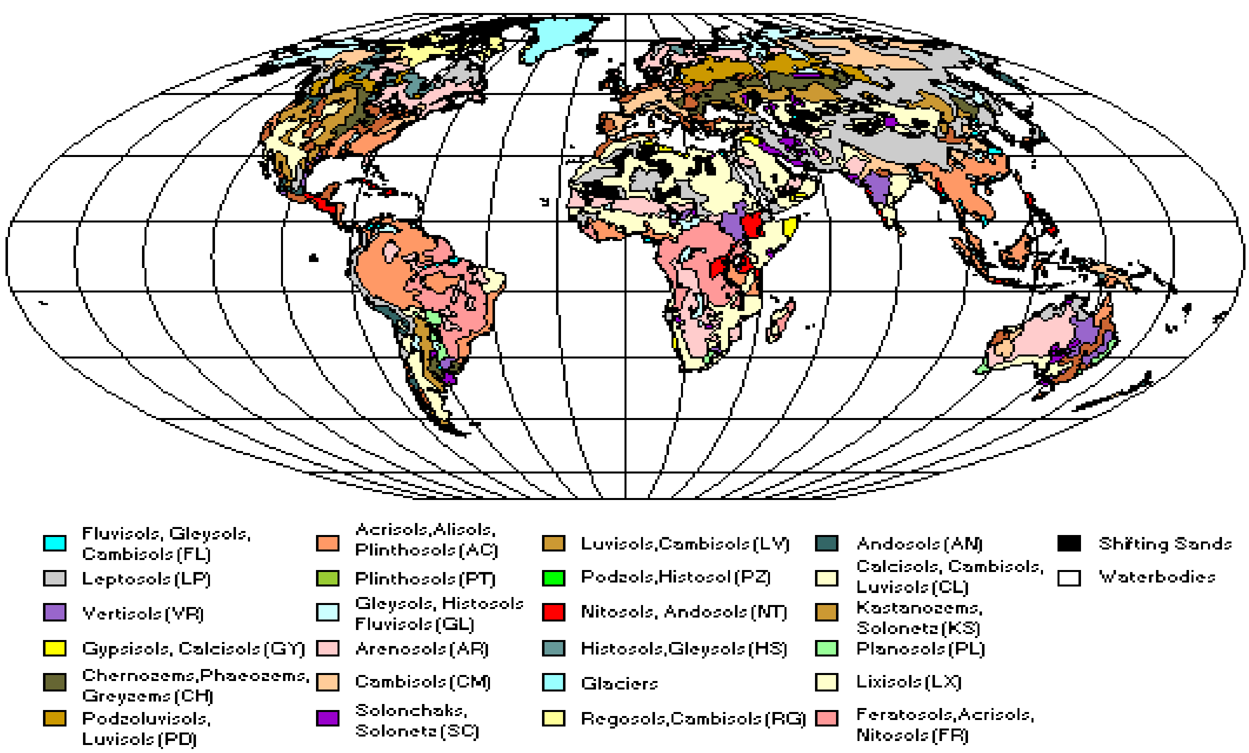

Figure 3-4. Major soil types worldwide.

3.3 BIOGENICS (SOILS)

The lack of a method to estimate ammonia emissions from soil is probably the most significant information gap that exists for ammonia inventories in general. Information and data are sparse, ambiguous, and/or uncertain. Ammonia emissions from soil, where they have been estimated, can dominate an inventory. As illustrated in **Figure 3-5**, soil-related ammonia emissions computed in the CMU tool represent almost 50% of all ammonia emissions. Most research conducted in recent years has shown that, at a given time, soil/plant canopy systems can act either as sources or sinks of ammonia (Roe and Mansell, 2001). For that reason among others, existing soil emission factors are highly uncertain.

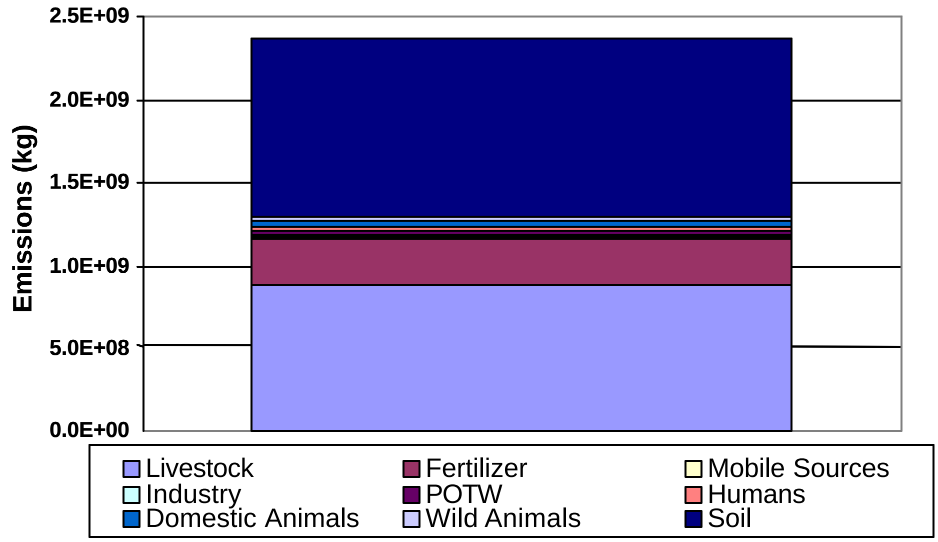

Figure 3-5. Ammonia emissions by source category for the LADCO and surrounding states computed using the original CMU emission factors and activity data.

Table 3-4 lists emission factors as alternatives to those used by the CMU tool. The information in Table 3-4 comes from a literature review by Corsi et al. (2000) and from follow-up measurements made in Texas (Corsi et al. 2002). The recommended emission factors appear to be the most appropriate for application in the United States. We recommend that LADCO use a seasonal allocation profile reported for Texas (Corsi et al., 2002) (see **Table 3-5**) but apply the springtime emissions rate—the lowest that Corsi reported—to better represent winter conditions in the LADCO area (see **Figure 3-6**).

Table 3-4. Biogenic soil emission factors.

Soil Type	Emission Factor (kg/km^2-yr)	Comment
Bare soil	370	Acknowledgement of available, though highly uncertain, information
Desert scrub	6.7	Consistent with recent measurements in Chihuahuan desert, similar to Texas
Grassland	40	Apparently consistent with 1997 measurement data in the western United States
Pasture	550	Median value from a variety of sources, which included measurements in United States, Australia, and Europe
Rangeland	14	Consistent with measurements in United States
Scrubland	100	Acknowledgement of available, though highly uncertain, information
Urban land area	400	Acknowledgement of available, though highly uncertain, information
Lawn surface	370	Acknowledgement of available, though highly uncertain, information
Oak forest	1.6	Consistent with recent measurements in Texas
Pine forest	1.1	Consistent with recent measurements in Texas
Other coniferous forests	40	Acknowledgement of available, though highly uncertain, information
Temperate forest and woodland and shrubland	400	Acknowledgement of available, though highly uncertain, information

Table 3-5. Biogenic soil emission seasonal allocation.

Emissions Source	Recommendation	Comment
All biogenics	In the short-term, apply seasonal profiles based on the report by Corsi et al. (2002). Longer-term, investigate use of the NASA Ames Ammonia Soil Emission Model <http://geo.arc.nasa.gov/sge/casa/>, and apply only after demonstration of reasonableness.	Better representation of real-world conditions.

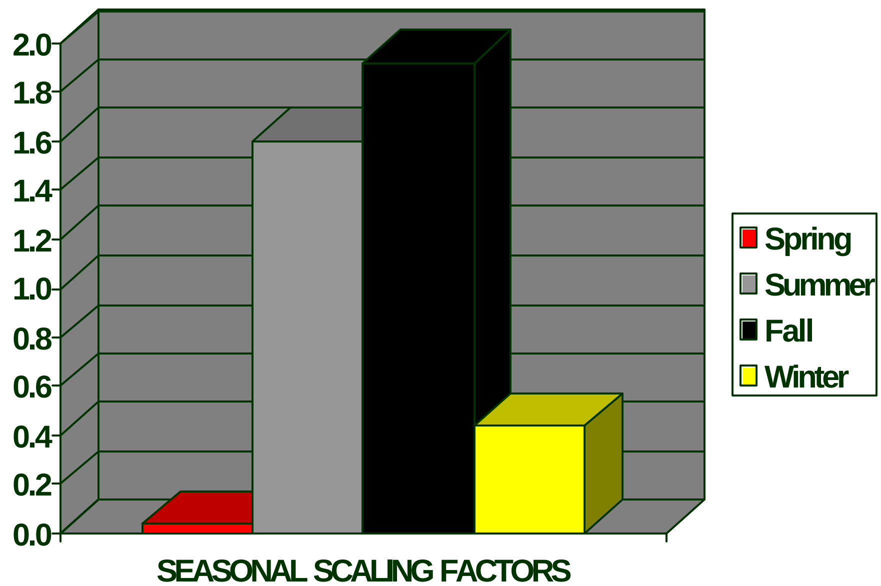

Figure 3-6. STI recommends the use of Texas "spring-like" levels for winter in the LADCO states.

As noted earlier, STI remains concerned because the Corsi et al., 2000, and Corsi et al., 2002, emission factors represent measurements at a point in time (apparently mostly in summer); therefore, they may not represent annual emission factors well. Adjusting these point-in-time local emission factors to represent annual conditions is an important task that should be undertaken (see Appendix A). As illustrated in Figure 3-6, summer emissions are thought to be twice as large as annual average emissions in Texas. Thus, the Corsi-recommended emission factors for soil likely need to be reduced by a factor of 2 to reflect annual average emission rates.

The CMU tool multiplies the biogenic soil emission factor for a land-use category by the area occupied by the land-use category within the county or state of interest. For the LADCO and surrounding states of interest, the total area of each state used in the CMU tool differs from other reference sources (see **Table 3-6**). Although not large on a percentage basis, the discrepancies of land area should be resolved because the impact of miscalculated emissions could be large.

Table 3-6. Area (square miles) of the LADCO and surrounding states.*

State	CMU Tool	Total Area (Land Area) http://www.50states.com/
Illinois	56,298	57,918 (55,593)
Indiana	36,400	36,420 (35,870)
Michigan	57,898	96,810 (56,809)
Ohio	41,193	44,828 (40,953)
Wisconsin	56,088	65,503 (54,314)
Iowa	56,257	56,276 (55,875)
Minnesota	84,517	86,943 (79,617)
Missouri	69,832	69,709 (68,898)

* Numbers in parenthesis are the surface area that is land.

Lastly, *we recommend further disaggregation of the land-use soil categories that are used in the CMU tool*. For example, "cropland" and "pasture" land-use categories are combined in a single land-use category. This combination confounds the fact that the fertilizer emission factor recommended by all the literature (see Table 3-3) includes biogenic ammonia emissions that also arise from cropland. Until the biogenic soil contribution from cropland is removed from the fertilizer emission factor for cropland, the inclusion by the CMU tool of a separate biogenic emission factor from cropland is problematic. Similarly, if livestock waste is being applied to pastures in an area, it is inappropriate to include a separate biogenic emission source from pastures because the existing livestock emission factors (Table 3-1) already incorporate corresponding biogenic emissions. According to the European Environment Agency (2001), biogenic losses from pastures were considerably smaller (by a factor of 3) relative to total emissions from pastures grazed by cattle.

Because there are uncertainties about and shortcomings in the biogenic emission factors and until new research shows improvement in the state of knowledge in characterizing ammonia emissions from soils, we recommend that biogenic (natural) soil emissions not be incorporated by LADCO.

Other Rural Sources

Biomass combustion (prescribed burning, wildfires, agricultural burning) may be a significant source of ammonia. This source is being evaluated as part of an ongoing inventory study in the San Joaquin Valley, California.

4. SUMMARY AND CONCLUSIONS

STI reviewed available literature to provide recommendations for updated emission factors for use in the CMU ammonia model as well as alternative sources for current activity data. Different emission factors for several large source categories were identified. In particular, emission factors for livestock, fertilizers, mobile sources, and POTWs should be revised. If applied, these revisions will result in substantial changes to the overall ammonia emission inventory. The recommended revisions will likely have the following impacts:

- Decrease in cattle emissions by about 50%
- Decrease in pig emissions by about 30%
- Increase in fertilizer emissions by about 50%
- Increase in gasoline-powered on-road vehicle emissions by about a factor of 10

Sources for the requisite activity data are summarized in **Table 4-1**.

Table 4-1. Sources of activity data.

Source Category	Data	Data Source
Cattle and calves Pigs and hogs Other livestock	Annual average number of livestock	U.S. Department of Agriculture <http://www.nass.usda.gov:81/ipedb/>
Fertilizer	Fertilizer usage	Association of American Plant Food Control Officials <http://www.aapfco.org/>
Mobile sources	VMT	U. S. Department of Transportation <http://www.fhwa.dot.gov/ohim/hs97/hs97page.htm>

Profiles to reallocate average annual emissions to hourly seasonal emissions were also provided for major source categories as well as a default allocation profile for all sources combined (see Section 1, Tables 1-3 and 1-4).

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APPENDIX A

DEVELOPMENT OF ANNUAL EMISSION FACTORS

Adjusting point-in-time local emission factors to represent annual conditions is not a simple undertaking but should be considered. The following three-step procedure is one approach that can be taken to obtain an annual emission factor for any location from a point-in-time emission factor measured elsewhere:

1. An equation describing the variation in the ammonia emission factor with ambient conditions is needed (see for example Equation A-1):

$$\text{Ammonia Emission Factor Equation} \sim U^a T^b \qquad \textbf{(A-1)}$$

2. Collect measurements of ambient condition present during the emission factor measurement (e.g., temperature and wind speed by time of day).
3. Determine an annual emission factor by accounting for variations in a year of representative ambient meteorological data as shown in Equation A-2:

$$\text{Annual Ammonia Emission Factor} = |\text{Measured Ammonia Emission Factor} \times \{(S_i\, U^a_i\, T^b_i) / 8760\} / U^a_m\, T^b_m \qquad \textbf{(A-2)}$$

where the subscript $_i$ denotes an hourly measurement of wind speed (U) and temperature (T) at a site with a year (8760 hours) of data nearby the location where an annual emission factor is desired. The subscript $_a$ identifies the measured wind speed (U) and temperature (T) corresponding to the measured ammonia emission factor.

Future research efforts involving measurements of ammonia emission factors need to require the corresponding measurements of ambient conditions if progress is to be obtained in formulating (improved) annual emission factors.

APPENDIX B

CONFLICTING SEASONAL AND DIURNAL LIVESTOCK EMISSIONS DATA

This appendix provides further details of the wide variations in reported seasonal and diurnal livestock emissions, particularly for cattle and swine.

Pierce and Bender (1999) proposed a seasonally invariant allocation scheme for dairy cows as a first approximation because of the controlled conditions and relatively stable rates of nutrition intake by commercially raised animals. This proposed approach is supported by data collected by Oosthoeck et al. (1991) who determined that emissions from a dairy house remained nearly constant from January to June. However, other European studies (Hartung, 1991; Mannebeck and Oldenburg, 1991) measured a doubling of emissions when ambient temperatures increased from 0°C to 20°C.

Sherlock and Goh (1984) determined mean volatilization rates for urine and urea-treated livestock pasture plots in New Zealand throughout the year. Averaging their results for urea and urine-treated soils yields seasonal adjustment factors of 24% of annual emissions in the summer, 32% in each of spring and fall, and 12% in winter.

Daily variations in ammonia emission from a naturally ventilated dairy-housing unit was studied by Zhu et al. (2000a, 2000b) as reported by Iowa State University and the University of Iowa Study Group (2002). During one day of monitoring, a consistent 1 ppm of ammonia concentration inside the housing unit was measured with a resulting emission rate averaging 4 ug NH3/m^2-s (0.35 g NH_3/m^2-day).

A theoretical equation developed by Russell and Cass in 1986 as reported by Coe et al. (1998) predicts diurnal emission changes from meteorological variations (Sadeghi and Dickson, 1992). The Russell and Cass equation (Equation B-1) relates hourly ammonia emission rates to temperature and wind speed as follows:

$$E_i \propto [2.36^{(T_i - 273)/10}] V_i^{0.8} A \qquad \textbf{(B-1)}$$

where

E_i = emission rate at hour i from animal waste decomposition

A = daily total emission rate for ammonia from animal waste = $\sum_{i=1}^{24} E_i$

T_i = ambient temperature in degrees Kelvin at hour i

V_i = wind speed in meters per second (m/s) at hour i (a minimum wind speed of 0.1 m/s is assumed)

A number of studies (Jacobson et al., 1996; Harris and Thompson, 1998; Todd, 1999; and Robarge et al., 2000) were examined to determine an overall seasonal emission profile for swine. However, the research which was all based on ambient air concentrations of ammonia near swine farms was contradictory. Some researchers found the highest ammonia levels in winter while others found the lowest levels during the winter.

Aneja et al. (2000) as reported by Iowa State University and the University of Iowa Study Group (2002) studied the seasonal variations in ammonia-nitrogen flux from an anaerobic lagoon in North Carolina and found maximum ammonia emissions during the summer (4017 µg N/m^2-min) with minimum levels in the winter (305 µg N/m^2-min). Mild weather emissions ranged from 844 (fall) to 1706 (spring) µg N/m^2-min. Equation B-2 was used to correlate these emission rates with lagoon surface temperature (measured 15 cm below the lagoon surface):

$$\mathrm{Log10(NH3\text{-}N)} = 2.1 + 0.048{*}T \qquad \textbf{(B-2)}$$

where the emission rate of ammonia is reported in terms of nitrogen (NH_3-N) and the emission rate units are µg N/m^2-min. T is the lagoon surface temperature in degrees Celsius. However, it is unfortunate that no attempt was made to relate the lagoon surface temperature to ambient air conditions; thus, this equation is not immediately practical to use for LADCO's interim needs.

Aarnink (1997) found that emissions from pig houses varied diurnally. Ammonia emissions from houses with rearing pigs and houses with fattening pigs had higher emissions during the day than during the night: +10% for rearing pigs and +7% for fattening pigs. For rearing pigs, emissions peaked in the morning, but for fattening pigs they peaked in the afternoon. Aarnink (1997) suggests that this seems to be related to the behavior of the animals. Also, Harris (2001) noted a significant diurnal cycle in ammonia emissions.

Ammonia emissions arising from a livestock lagoon were determined by Mount et al. (2001) to increase diurnally by a factor of 2.5 for an 11^{o}C increase in ambient temperature. While this study illustrates that emissions from lagoons are temperature-dependent, it lacks depth of analysis. Both diurnal wind speed and atmospheric stability variations affect emissions. Since it is not clear what the winds or temperature profile were during these measurements, it is not known whether this finding is representative of "typical" ambient conditions or represent an anomaly.

Cohen and Ryan (1985) provide a theoretical equation, supporting model development documentation, and validation of observations illustrating the role of water temperature and wind speed on emissions from a lagoon. Fundamentally, the Cohen and Ryan (1985) model for emissions from lagoons should provide a better longer-term solution for LADCO to estimate the diurnal ammonia emission profile from lagoons than the empirical data of Mount et al. (2001), because the model accounts for emission differences associated with changes in wind speed, temperature, and stability.