Quality Improvement for Ammonia Emission Inventory

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ABSTRACT

Ammonia gas reactions in the atmosphere are a significant source of PM2.5. Due to the increasing concentration of PM2.5 in atmosphere, the need for improved ammonia emission inventories has become an increasingly important air quality issue. A comparison is shown of ammonia inventory data from CMU (Carnegie Mellon University) based on the years 1996 and 1997, and NET99 (EPA's National Emission Trends database for 1999). The CMU data include most source categories but not soil and vegetation. NET99 appears to underestimate emissions for area and point sources in some southeast states, and it lacks details about emission categories such as a breakdown of livestock and domestic sources. NET99 also has incomplete activity information that is needed to provide details about sub-source categories in a county. This study presents the improved ammonia emission estimates that were developed for each county in Tennessee. This study produced ammonia emission estimates for ten source categories and fifty-four sub-source categories for the State of Tennessee on a counties basis for the year 1999.

Results show that livestock is a major source of total ammonia emissions in Tennessee, responsible for emitting 88,000 tons per year, with beef cows emitting 45,000 tons of ammonia emissions and forming the biggest sub-source category within the livestock category. Even though soil may be an uncertain source category of ammonia emissions and some double counting of emissions may occur in the fertilizer source category, soil was estimated to emit 61,000 tons of ammonia emissions as the second highest contributor. In some urban counties in Tennessee, mobile sources were the largest source of ammonia emissions. The study includes recommendations for the use of spatial surrogates to allocate emissions for area sources, and temporal profiles for allocating emissions by season and time of day.

INTRODUCTION

Gaseous ammonia is an important precursor pollutant that contributes to the formation of $PM_{2.5}$ (particulate matter with an aerodynamic diameter less than 2.5μm). Ammonia gas generated from various sources reacts chemically with nitric and sulfuric acid in the atmosphere. The chemical reactions form $PM_{2.5}$ that has effects on human health and visibility. Because of this, an ammonia emission inventory by county for the State of Tennessee is needed for modeling $PM_{2.5}$ formation.

The objective of the study is to develop an improved ammonia emission inventory by county for the State of Tennessee for the base year of 1999. Previous studies by CMU (Carnegie Mellon University) have generated an ammonia emission inventory for the State of Tennessee. Ammonia emissions in the CMU study (CMU, 2001) based on the years of 1995, 1996, and 1997 were found to lack information about several emission source categories such as human, soil, and mobile sources. NET99 (EPA's National Emission Trends, 1999), which also contains NH_3 inventory, was found to contain an incomplete emission inventory by county and lacked details about sub-source categories. To improve the quality of the ammonia inventory for model $PM_{2.5}$ formation for the year of 1999 for the state region, this study presents 10 source categories and 54 sub-source categories for improved 1999 ammonia emission inventory estimates by county for the State of Tennessee. Spatial and temporal allocations for sub-source categories for modeling $PM_{2.5}$ formation are also presented.

BACKGROUND

Ammonia is present even in unpolluted air as a result of natural biochemical and chemical processes. High concentrations of ammonia gas in the atmosphere are generally indicative of accidental release of the gas. Ammonia is removed from the atmosphere by its affinity for water and by its action as a base. It is a key species in the formation and neutralization of nitrate and sulfate aerosols in polluted atmosphere (Manahan, 1999).

Atmospheric chemical reactions convert NOx to nitric acid, inorganic nitrate salts, organic nitrates, and peroxyacetyl nitrate. The principal reactive nitrogen oxide species in the troposphere are NO, NO_2, and HNO_3. Nitrogen dioxide is a very reactive and significant species in the atmosphere and is ultimately removed from the atmosphere as nitric acid, nitrates as organic nitrogen. In the stratosphere, nitrogen dioxide reacts with hydroxyl radicals to produce nitric acid. In this region, the nitric acid can also be destroyed by hydroxyl radicals, or by a photochemical reaction, so that HNO_3 serves as atemporary sink for NO_2 in the stratosphere. Nitric acid produced from NO_2 is removed as precipitation, or reacts with bases (ammonia, particulate lime) to produce particulate nitrates (Manahan, 1999).

Ammonia gases emitted from a wide range of human-generated and natural sources react chemically with nitric acid and sulfuric acid in the atmosphere. An important role of ammonia in the atmosphere is neutralizing compounds such as nitric and sulfuric acids, formed as products of the atmospheric oxidation of NOx and SO_2 emissions, respectively (Kean et al., 2000). The summary equilibriums are as follows.

$$NH_{3\,(g)} + HNO_{3\,(g)} \rightarrow NH_4NO_{3\,(s)}$$

$$NH_{3\,(g)} + H_2SO_{4\,(g)} \rightarrow NH_4HSO_{4\,(s)}$$

The chemical reactions contribute to form $PM_{2.5}$ (particulate matter with an aerodynamic diameter less than 2.5 um). Ammonium nitrate and sulfate contribute significantly to fine particle mass and visibility problems (Chow et al., 1994;Larson et al., 1988). The transport of nitric acid is aided by the formation and condensation of ammonium nitrate on accumulation mode particles. Furthermore, dry and wet deposition of ammonia and ammonium is an important source of nutrients to some ecosystems (Pearson and Stewart, 1993). In the situation of high ammonia and nitric acid concentrations and low sulfate concentrations in the atmosphere, gaseous ammonia can react to form ammonium nitrate. In the presence of sulfate concentrations, gaseous ammonia can react to form ammonium sulfate. Gaseous SO_2 reacts with the hydroxyl radical resulting in the formation of sulfuric acid. Whether reacted with nitric or sulfate acid, the ammonium ion (NH_4^+) is often regarded to be an important factor assoc Global ammonia emissions were estimated to be 45 million metric tons per year, with approximately two- thirds of this total being attributed to human-generated sources. Almost one-half of total ammonia emissions were attributed to animal husbandry. Ammonia is the most influential gaseous base in the atmosphere and a main neutralizing factor for atmospheric acids. The NH_3 in the atmosphere may control the acidity of precipitation (Dentener and Crutzen, 1994).

Ammonia also interacts with regional and global atmospheric sulfur cycles, thus having a potential impact on regional visibility and global warming. $PM_{2.5}$ as the secondary fine particulate matter that is formed by reactions involving ammonia are a growing concern in terms of impacts on human health (Bouwman et al., 1997). Neutralization products such as ammonium bisulfate, ammonium sulfate, and ammonium nitrate are known to be significant constituents of atmospheric aerosols, including cloud condensation nuclei (Dawson, 1977).

CMU INVENTORY

The CMU study (CMU, 2001) generated ammonia emission estimates for the U.S. on a state-by-state basis. Figure 1 shows a pie chart from the CMU inventory (CMU, 2001) for the State of Tennessee. There were 9 source categories and 36 sub-source categories for annual emission estimates in the inventory. Table 1 provides a list of ammonia source categories and sub-source categories used in the CMU inventory. (ArcGIS 8.1, 2001). Table 2 presents ammonia emission estimates for the 36 sub-source categories by county in Tennessee.

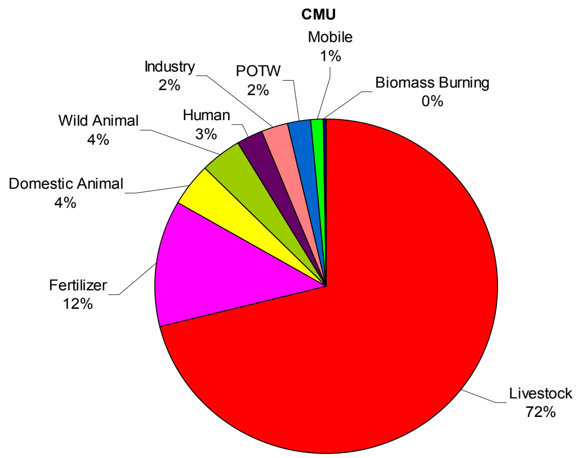


Figure 1. Pie Chart of Ammonia Emission Estimates from CMU Inventory in Tennessee by Source

Table 1. Source, Sub-Source Category, and Emission Factors in CMU Inventory

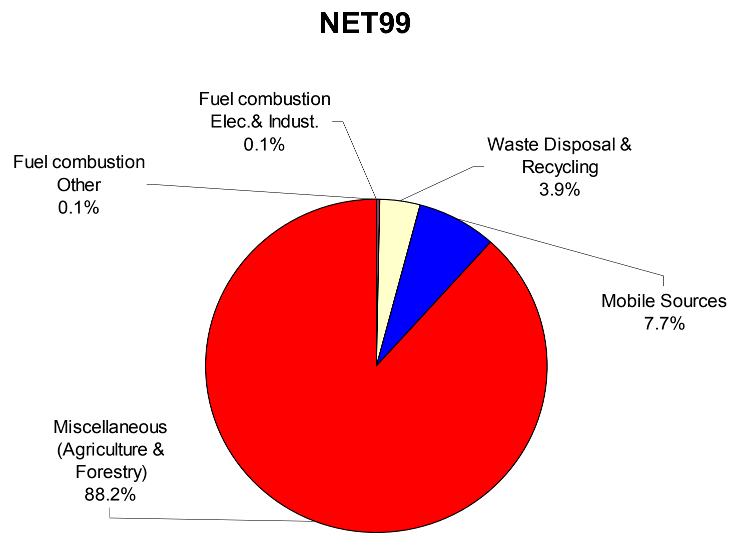
Source Category	Sub-Source Category	Emission Factor	Units
Livestock	Milk Cows	39.72	kgNH3/cow/yr
	Beef Cows	39.72	kgNH3/cow/yr
	Heifers and Heifer Calves	13.04	kgNH3/heifer/yr
	Steers,Steer Calves,Bulls, and Bull Calves	8.22	kgNH3/steer/yr
	Hogs and Pigs	9.2	kgNH3/pig/yr
	Horses and Ponies	12.2	kgNH3/horse/yr
	Sheep and Lambs	3.37	kgNH3/sheep/yr
	Angora Goats	6.4	kgNH3/goat/yr
	Milk Goats	6.4	kgNH3/goat/yr
	Pullets	0.269	kgNH3/pullet/yr
	Layers	0.598	kgNH3/Layer/yr
	Broilers	0.167	kgNh3/broiler/yr
	Turkeys	0.858	kgNH3/turkey/yr
	Geese	0.89	kgNH3/goose/yr
	Ducks	0.117	kgNH3/ducks
Human	Human	0.44	kgNH3/person/yr
Fertilizer	Anhydrous_Ammonia	0.01	percent N released to atmosphere
	Ammonium_Nitrate	0.02	percent N released to atmosphere
	Ammonium_Sulfate	0.08	percent N released to atmosphere
	Ammonium_Thiosulfate	0.025	percent N released to atmosphere
	Calcium_Ammonium_Nitrate	0.02	percent N released to atmosphere
	Nitrogen_Solutions	0.08	percent N released to atmosphere
	Urea	0.15	percent N released to atmosphere
	Diammonium_Phosphate	0.04	percent N released to atmosphere
	Monoammonium_Phosphate	0.04	percent N released to atmosphere
	Liquid_Ammonium_Polyphosphate	0.04	percent N released to atmosphere
	Potassium_Nitrate	0.02	percent N released to atmosphere
	Miscellaneous	0.15	percent N released to atmosphere
	Mix	0.04	percent N released to atmosphere
Mobile	Cars	1.35E-05	kgNH3/VMT/yr
	Trucks	6.83E-05	kgNH3/VMT/yr
POTW			
Industry			
Domestic Animals	Dogs	2.18	kgNH3/dog/yr
	Cats	0.69	kgNH3/cat/yr
Wild Animals	Deers	4.54	kgNH3/deer/yr
	Bears	4.54	kgNH3/bear/yr
Biomass Burning	Fire_NH3	0.012	moleNH3/moleCO
	Fire_CO	140	lbsCO/ton wood burned
9 Source Category	36 Sub-Source Category		

Table 2. Ammonia Emission Summary for Tennessee in CMU Inventory

Source Category	Sub-Source Category	Ammonia Emissions	Units
Livestock	Milk Cows	4,903.13	ton/yr
	Beef Cows	45,516.89	ton/yr
	Heifers and Heifer Calves	7,200.67	ton/yr
	Steers,Steer Calves,Bulls, and Bull Calves	4,466.08	ton/yr
	Hogs and Pigs	3,263.52	ton/yr
	Horses and Ponies	1,197.12	ton/yr
	Sheep and Lambs	51.16	ton/yr
	Angora Goats	5.63	ton/yr
	Milk Goats	27.37	ton/yr
	Pullets	244.58	ton/yr
	Layers	1,090.38	ton/yr
	Broilers	3,281.72	ton/yr
	Turkeys	2.37	ton/yr
	Geese	2.82	ton/yr
	Ducks	1.31	ton/yr
	Total	**71,254.74**	ton/yr
Human	Human	**2,606.08**	ton/yr
Fertilizer	Anhydrous_Ammonia	763.05	ton/yr
	Ammonium_Nitrate	4,002.50	ton/yr
	Ammonium_Sulfate	34.19	ton/yr
	Ammonium_Thiosulfate	0.23	ton/yr
	Nitrogen_Solutions	947.01	ton/yr
	Urea	2,612.26	ton/yr
	Diammonium_Phosphate	2121.9	ton/yr
	Monoammonium_Phosphate	13.2	ton/yr
	Liquid_Ammonium_Polyphosphate	26.55	ton/yr
	Potassium_Nitrate	2.96	ton/yr
	Miscellaneous	159.31	ton/yr
	Mix	1,442.22	ton/yr
	Total	**12,125.38**	ton/yr
Mobile	Cars	749.88	ton/yr
	Trucks	381.38	ton/yr
	Total	**1131.26**	ton/yr
POTW		**2321.77**	ton/yr
Industry		**2467.59**	ton/yr
Domestic Animals	Dogs	3477.20	ton/yr
	Cats	921.84	ton/yr
	Total	**4399.04**	ton/yr
Wild Animals	Deers	3703.12	ton/yr
	Bears	5.00	ton/yr
	Total	**3,708.12**	ton/yr
Biomass Burning	Fire_NH3	**202.18**	ton/yr
Total		**100,216.16**	ton/yr
Total		**91,013,811.11**	kg/year

NET 99 TIER (EPA's National Emission Trends Tier, 1999)

The search began with a review of ammonia emissions that had previously been reported in the US Environmental Protection Agency's National Emission Trends Tier Report for the year of 1999 (NET 99 Tier). The NET 99 Tier report provided emission estimates for ammonia sources in Tennessee. However, it was found to underestimate ammonia emissions for point sources and it had no breakdown of area sources such as livestock, fertilizer, and soil sources when compared to the CMU inventory. The NET99 Tier inventory only classified 3 source categories (area, mobile and point sources) with SCCs (Source Classification Codes). The SCCs in each source category (area, point and mobile) were not found to be complete. For example, the SCCs in the area source category only included sub-source categories of on-highway and off-highway vehicles (diesel or gasoline), railroad equipment, construction and mining equipment. Sub-source categories for livestock and fertilizer, typically considered to be the major sources in an ammonia emission inventory, were not even included in the NET99 inventory. Figure 2 shows a pie chart from NET99 ammonia emission inventory for the State of Tennessee. It underestimates emissions from point sources showing only 89 tons per year. Table 3 presents ammonia emission estimates by county for 3 source categories in Tennessee showing a total of 82,260 tons per year compared to over 100,000 tons per year for the CMU inventory. The unit of emission estimates for each sub-source category used in this



study is short tons per year.

Figure 2. Pie Chart of Ammonia Emissions in NET99 for Tennessee

Table 3. Ammonia Emissions by Source for Tennessee

Source Category	Ammonia Emissions	Units
Area	75,811	Ton/yr
Mobile	6,360	Ton/yr
Point	89	Ton/yr
Total	82,260	Ton/yr

METHODOLOGY

Estimate of ammonia emissions for each sub-source category is provided based on the equation shown below:

E = EF x AL

where:

E = Emission Estimates for sub-source categories
EF = Emission Factor for sub-source categories
AL = Activity Level for sub-source categories

Table 4 shows ten source categories and fifty-four sub-source categories used in this study. Based on the literature review and reports by CMU (CMU, 2001), it was determined that the ammonia emission inventory should consist of 10 source categories and 54 sub-source categories as shown in Table 4. Emission factor, activity data, and activity data source are tabulated by each sub-source category.

Table 4. Source Categories and Sub-Source Categories Used in This Study

Source Categories	Sub-Source Categories
Livestock	Cattle&Calves, Milk Cows, Beef Cows, Hog & Pig, Broilers, Layers(>1 year), Layers(<1year), Pullets(<13 weeks), Pullets(>13 weeks), Other Chickens, Horses & Ponies composite, Sheep & Lambs Composite, Goats, Ducks, Geese, Turkeys
Soil	Urban, Forest, Wetland, Agricultural land
Fertilizer	Anhydrous Ammonia, Nitrogen Solutions, Urea, Ammonium Nitrate, Ammonium Sulfate, Ammonium Thiosulfate, Other Straight Nitrogen, Ammonium Phosphates, N-P-K
Mobile Sources	LDGV, LDGT12, LDGT34, HDGV, MC, LDDV, LDDT, HDDV
	NonRoad Gasoline, NonRoad Diesel, Aircraft, Marine Vessels, Railroads
Human Sources	Human Perspiration, Human Respiration, Untreated Human Waste, Cigatette, Household Ammonia Use, Cloth Diapers, Disposal Diapers, Homeless
Biomass Burning	None
Industry	None
POTW	None
Wild Animal	Bear, Deer
Domestic Animal	Dogs, Cats

Note : 10 Source Categories 54 Sub-Source Categories

EMISSION FACTORS

Estimates of ammonia emissions depend on emission factors. In particular, it is believed that ammonia emissions from livestock and fertilizer are significant parts of total NH_3 emissions. Therefore it is important to use the most recent and accurate emission factors for estimating NH_3 emissions.

Recent NH_3 emission factor research in Europe, and specifically in the Netherlands, has focused on increasing the accuracy of NH_3 emission factors for livestock and fertilizer. In 1992, a report on the NH_3 emissions in Europe was published by Asman, 1992. The report incorporated research developed in the Netherlands through around 1990. The emission factors presented by Asman, 1992 are the most recent and accurate emission factors for livestock and fertilizer. In 1994, a final report of development and selection of ammonia emission factors was published by Battye et al., 1994 which included the emission factors.

Emission factors affecting ammonia emissions include: animal age, kind, weight, animal housing system, nitrogen content of feed, manure storage practices, the way of applying the manure or fertilizer, time between spreading and plowing, meteorological conditions, and soil properties. However, data for many of these parameters were not available. For this study, composite emission factors obtained from Battye et al., 1994 for some sub-source categories are chosen to estimate more accurate ammonia emissions from livestock and fertilizer. Emission factors from other sources such as soil, human, and domestic animals were obtained from the Corsi et al., 2000 report. This study presents emission factors used for sub-source categories, a quality rating for the emission factors taken from Battye et al., 1994, and activity levels (population) to estimate ammonia emissions for the year 1999.

ACTIVITY DATA

Estimates of ammonia emissions also depend on activity data. It is quite hard to collect accurate and detailed activity data by county for each sub-source category. Furthermore, activity data (populations) of livestock classified by weight groups were not available. Activity data for ducks, geese, turkeys, wild animals, and POTWs (Publicly Owned Treatment Works) were obtained for the years 1996 and 1997. They were not available for the year 1999. Because mink, fox, and rabbit data were not available, they were not included in this study. This study presents a wide range of information sources, which were used to obtain necessary activity data for 1999.

SUMMARY OF RESULTS OF STATEWIDE AMMONIA EMISSIONS BY SOURCE

This section summarizes the results of statewide ammonia emission estimates for the year 1999 for the State of Tennessee. Figure 3 shows the statewide ammonia emission estimates and percentages for each primary source category and sub-source category. Ammonia emission estimates for sub-source categories are listed in Table 5 for each primary source category and sub-source category for 1999.

Livestock was the largest major source, responsible for 48% of the total ammonia emissions, with beef cows accounting for 51.3% of the livestock ammonia emissions. Soils were the second largest major source, accounting for 33% of the total ammonia emissions. Fertilizers were estimated to account for 6% of the total ammonia emissions. Mobile sources were estimated to contribute 4% of the total ammonia emissions, with

LDGV (Light- Duty Gasoline Vehicles) accounting for 57% of the mobile emissions. The following categories were each estimated to contribute 2% of the inventory: wild animals, domestic animals, industrial sources according to the TRI (TRI, 1999), and human sources. POTW were estimated to contribute 1% of the inventory. Biomass burning played the smallest role in ammonia emissions, comprising less than 0.1% of the total. Figure 4 shows the ammonia emission distributions by county by source category.

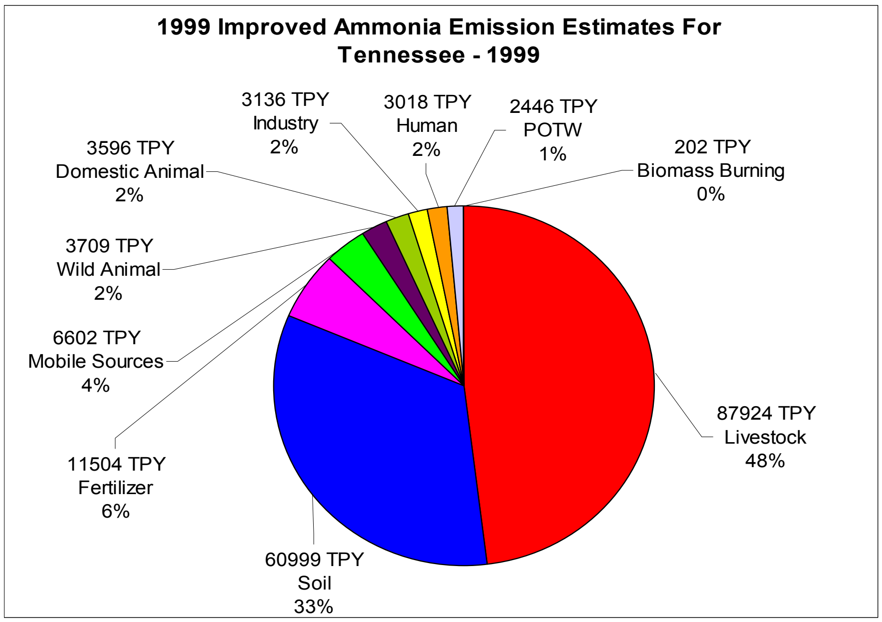


Figure 3. Ammonia Emission Estimates and Percentages for Source Categories

Table 5. Ammonia Emission Estimates for Sub-Source Categories in Tennessee

Source Category	Sub-Source Category	Emissions (tons/year)
Livestock	Milk Cows	4378.64
	Beef Cows	45097.31
	Cattle & Calves	26505.07
	Hog & Pig	2504.64
	Broilers	4633.00
	Layers (>1 yr)	425.00
	Layers (<1 yr)	849.9
	Pullets (>13 Weeks)	292.34
	Pullets (<13 Weeks)	219.99
	Other Chickens	90.11
	Horses	2650.62
	Sheeps	47.82
	Goats	223.13
	Ducks	1.31
	Geese	2.79
	Turkeys	2.37
	Total	**87924.04**
Soil	Urban land	1588.94
	Forest land	21932.94
	Wetland	484.77
	Agricultural land	36992.53
	Total	**60999.18**
Fertilizer	Anhydrous Ammonia	131.87
	Nitrogen Solutions	508.45
	Urea	6164.18
	Ammonium Nitrate	1257.69
	Ammonium Sulfate	110.68
	Ammonium Thiosulfate	2.43
	Other Straight Nitrogen	1038.76
	Ammonium Phosphates	1290.58
	N-P-K	999.23
	Total	**11503.87**
On-Road Mobile Sources	LDGV	3709.62
	LTGT12	1857.49
	LTGT34	615.29
	HDGV	52.58
	MC	1.52
	LDDV	0.22
	LDDT	0.18
	HDDV	155.61
Nonroad Mobile Sources	Nonroad Gasoline	10.00
	Nonroad Diesel	60.00
	Aircraft	92.00
	Marine Vessels	35.00
	Railroads	13.00
	Total	**6602.51**
Wild Animals	BlackBears	5.00
	Deer	3703.12
	Total	**3708.13**
Domestic Animals	Dogs	2655.32
	Cats	941.17
	Total	**3596.49**
Industry		**3135.69**
Human	Human Perspiration	1208.91
	Human Respiration	1208.91
	Untreated Human Waste	120.89
	Cigarette Smoking	0.006
	Household Ammonia Use	181.34
	Cloth Diapes	100.40
	Disposal Diapers	46.64
	Homeless	151.11
	Total	**3018.22**
Biomass Burning		**202.18**
POTW		**2446**
Total Ammonia Emissions		**183136.12**

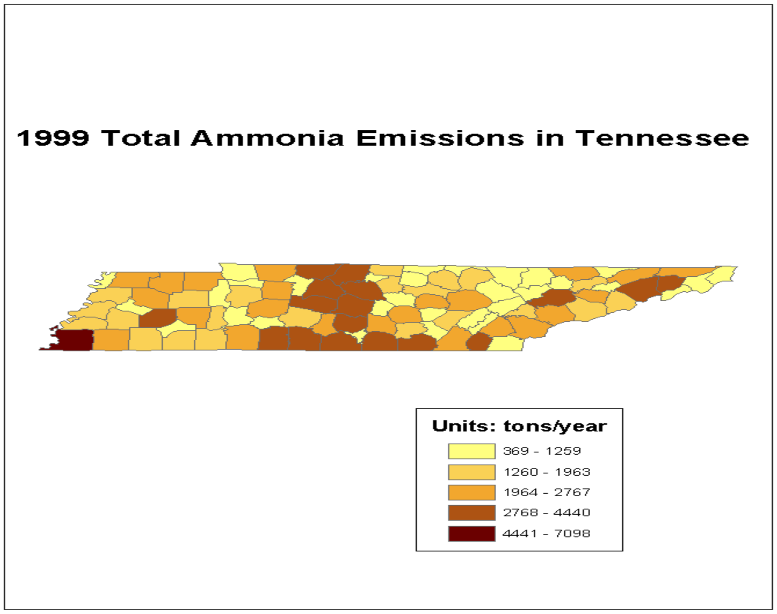


Figure 4. Total Ammonia Emission Estimates for 1999

CONCLUSIONS AND RECOMMENDATIONS

This study has estimated annual ammonia emissions from 54 sub-source categories on a county basis for the state of Tennessee. The base year of the study was 1999. Ammonia emission estimations were made separately for all 95 counties in Tennessee. The conclusions in this study are listed below:

- Livestock is the major source of total ammonia emissions in Tennessee, responsible for emitting 87,924 tons per year, with beef cows emitting 45,097 tons of ammonia emissions, forming the biggest sub-source category within the livestock category. Even though soils may be an uncertain source category of ammonia emissions, soils emitted 60,999 tons of ammonia emissions as the second largest contributor.
- Yearly ammonia emissions from fertilizers were considered in this study. However, monthly fertilizer ammonia emissions are likely to be more accurate than yearly averages for the temporal profile. Annual fertilizer usage was used with temporal default plots to allocate ammonia emissions from fertilizer. It would be better to have monthly application rates so the default file would not be needed.
- Fertilizer ammonia emissions were found to be the third largest contributor, estimated at 11,504 tons per year, with urea emitting 6,164 tons of ammonia emissions for Tennessee in 1999.
- In Knox, Davidson, Hamilton, and Shelby counties, the total emission magnitudes from soil sources were larger than those of livestock sources.

As a result of the study, ammonia emission estimates are available on a county-level basis and can be used for modeling $PM_{2.5}$ for Tennessee. The results and approach should make contributions to the literatures. In particular, ammonia emission estimates from area sources can be useful for modeling $PM_{2.5}$ as input data. In addition, the spatial surrogates for area sources are identified in this study for modeling. Additional research is needed to more accurately estimate ammonia emissions, including the development of emission factors and the development of more refined temporal allocations for fertilizers and livestock sources.

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