An Improved Ammonia Inventory for the WRAP Domain

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ABSTRACT

Recent advances in the understanding of the health impacts of particulate pollution and the important role ammonia (NH3) emissions play in the formation of secondary particulate matter (PM) has spawned a great deal of new research into ammonia emissions. Major sources of NH3 emissions include livestock operations, fertilizer use, waste management, mobile sources, industrial point sources, and various biological sources including human respiration, wild animals, and soil microbial processes. For each of these source categories there remain large uncertainties in the magnitude of emissions, the diurnal and seasonal variation, and the spatial distribution. Uncertainty in NH3 emissions is a key source of uncertainty in the formation of sulfate and nitrate aerosols. Thus, development of improved NH3 emissions inventories is essential for modeling the formation of fine PM, regional haze, and for developing effective plans to mitigate visibility impairment at National Parks, Forests and Wilderness Areas.

Significant improvements have been made in the understanding of ammonia emissions since the development of the current 1996 National Emissions Inventory (NEI) that was used in the Western Regional Air Partnership (WRAP) visibility modeling to meet Clean Air Act Section 309 requirements. Particularly, the temporal dependence of ammonia emissions on environmental parameters has been the focus of several recent research efforts. The WRAP has provided funding to the Regional Modeling Center (RMC) to develop an improved NH3 emissions inventory for the WRAP States and tribes to use in Clean Air Act Section 308 modeling.

A secondary goal of the project is the development of an ammonia-capable geographic information system (GIS) based emission modeling system. Emission estimates for many ammonia source categories are intimately linked to land use/land cover (LULC) characteristics and environmental variables. Hence, a modeling system which combines the GIS data on LULC distributions with LULC-specific emission factors and activity indicators can calculate gridded emissions in a single processing step and is ideally suited to development within a GIS framework. A GIS-based emission model was developed to provide an accurate and efficient tool for inventory processing, particularly the spatial allocation and gridding of the emission inventory. The emission model uses Arc/INFO GIS software and makes use of high resolution LULC data for the development of gridding surrogates. The user-friendly, menu-driven graphical user interface (GUI) facilitates efficient inventory generation, revisions and future updates with minimal processing effort and resources.

INTRODUCTION

Significant improvements have been made in the understanding of ammonia emission since the development of the existing inventory used for WRAP modeling. Additionally, more data has become available to help accurately allocate these emissions spatially. A recent study by Chinkin et al.[1] (2003) on ammonia emission inventory improvements for the Lake Michigan Air Director's Consortium (LADCO) provides a recent comprehensive review of available ammonia emission factors. Potter et al.[2] (2001), an inventory of fertilizer and soil ammonia emissions for the State of California, provides methodologies for estimating and distributing ammonia emissions from these sources based on environmental parameters. These two documents form the basis for this inventory effort. The ammonia emissions modeling system described in this document will allow for quick and easy development of an ammonia inventory for any spatial domain. The modeling system is to be populated the necessary data to produce a gridded inventory for the 2002 WRAP domain. The completed gridded inventory will be delivered with the modeling system. The development of the ammonia inventory and the GIS-based modeling system has been documented by Chitjian and Mansell (2003)[3,4].

Significant ammonia emissions include livestock operations, fertilizer use, waste management, mobile sources, industrial point sources, wild and prescribed fires, and various biological sources including human respiration, wild animals, and soil microbial processes. Of these source categories, the primary contributors to annual ammonia emissions in a national level include livestock operation, fertilizer use, soil microbial processes and, to a lesser extent, domestics sources. In the current inventory, only these source categories are considered. Ammonia emissions from mobile sources, industrial point sources and wild and prescribed fires are being developed by other WRAP Forums. The effects of environmental parameters on the emission factors and temporal variation are incorporated for livestock operations fertilizer use and native soils.

METHODS AND DATA SOURCES

Data sources, including activity data, emission factors, and environmental variables, and inventory development methods for each sources category included in the inventory are presented and discussed in this section.

Emission Factors

A summary of emission factors found in recent literature and the recommended values for use in the development of the WRAP ammonia inventory are presented in this section.

Livestock Operations

Battye et al.[5] (2003) provides a comprehensive review of livestock ammonia emission factors reported by several studies published since 1994. Because many livestock ammonia emission factors are obtained from measurements made in Europe, the authors also provide a comparison of these factors to estimates determined from the Midwest Plan Service, a handbook that provides estimates of ammonia losses to the atmosphere from various waste storage and management practices. The authors recommend a dairy ammonia emission factor of 28 kg-NH_3/animal-year, consistent with the waste design handbook of 20-70 kg-NH_3/animal-year. These values are consistent with those developed by the EPA Office of Research and Development (ORD) for cattle, swine and poultry (EPA, 2002)[6]. The poultry emission factors recommended by Battye et al.[5], 0.28 kg-NH_3/animal-year for broilers and 0.37 kg-NH_3/animal-year are within the range determined from the handbook but significantly greater than the ORD factors. The swine emission factors recommended by Battye et al.[5] , 6.4 kg-NH_3/animal-year and 16.4 kg-NH_3/animal-year for finishing pigs and sow respectively, are very similar to the 6.8 kg-NH_3/animal-year emission factor recommended by ORD.

Pinder et al.[7], (2003) report dairy cow emission factors ranging from 13.1 to 55 kg NH_3 /cow /year. Emission factors are higher in the southern and western states, which the authors attribute to both warmer temperatures and more intensive practices. The authors use a semi- empirical model of ammonia emissions from a dairy farm. The results of this model are combined with a statistical National Practices Model to estimate dairy cow emission factors throughout the country.

Keener et al.[8] (2001) studied ammonia emissions at a large modern poultry facility using two different management practices; deep-pit and compost/belt systems. Based on testing in March, the authors estimated 0.573 kg-NH_3/animal-yr using a mass balance approach or 0.669 kg-NH_3/animal-yr using air flow and ambient concentrations for the deep-pit system and 0.152 kg-NH_3/animal-yr using a mass balance approach or 0.531 kg-NH_3/animal-yr using air flow and ambient concentrations for the belt/compost system. These emission factors are fairly consistent with those recommended by Battye et al.[5] but are much greater than those recommended by ORD.

Doorn et al.[9], (2002) provides an emission factor of 7 kg-NH_3/animal-yr based on extensive testing at "Farm 10" in North Carolina, a swine operation, and some follow–up testing at other swine farms in southern North Carolina. Doorn et al.[9] also provides a comparison to several other researchers' results in addition to those reviewed by Battye et al.[5].

The current inventory is based on the livestock emission factors recommended by Chinkin et al.[1] for dairy cows, beef cows, poultry and swine. These factors are well documented and consistent with recent measurements and estimates. Emission factors for horses and sheep were taken from Battye et al.[5] as these estimates considered more recent research. Table 1 presents the emission factors used in the 2002 WRAP inventory and a comparison of other livestock emissions factors found in recent literature/

Fertilizer Use

The European Environment Agency (EEA, 2001[10]) has developed emissions factors based on fertilizer type, soil type and climate as follows:

Group I Warm temperate countries with a large proportion of calcareous soils (e.g. Greece, Spain).
Group II Temperate and warm-temperate countries with some calcareous soils (or managed with soil pH >7), but with large areas of acidic soils (e.g. Italy, France, UK, Eire, Portugal, Belgium, Netherlands, Luxembourg.)
Group III Temperate and cool-temperate countries with largely acidic soils (e.g. Nordic countries, Germany, Switzerland, Austria).

Potter et al.[2], determined emission factors ranging from 4-6.5 % for surface applied fertilizers and 0-1 % for subsurface (or buried drip or micro-drip technologies.) There is no distinction made between different types of fertilizers The authors estimate percent application types that vary by crop and growing areas within California, but are generally evenly split between surface and subsurface. From this information, an overall emission factor of approximately 2%-4% is obtained. A comparison of fertilizer emission factors is shown in Table 2.

The model developed by Potter et al.[2] estimates ammonia emissions from fertilizers as a function of fertilizer management practices and several environmental variables. They noted that results from experiments indicate changes in pH (>7.5) had the most significant effect on ammonia emissions (Dewes, 1996)[11]. The model included a rule-based assignment of emission factors based on soil pH and application category. The emission factors expressed as a percent of N applied are 4.0, 5.5, 6.5 for soil pH below 7, between 7 and 8, and above 8 respectively, for surface application. This represents a 60% increase with elevated pH. The authors noted no difference in ammonia emissions with different soil pH with subsurface application.

The current inventory is based on the fertilizer emission factors cited by EEA (2002)[10] for Group II. Because of the importance of soil pH in determining ammonia emissions from fertilizers, the base emission factors are adjusted for soil pH. A scalar was developed to adjust the fertilizer specific emission factors for soil pH. The scalar was based on a linear regression of the values cited by Potter et al.[2]. This regression was normalized to the 4% emission factor at pH=6.5, yielding a normalized emission factor scalar of the form

$$\text{Scalar} = (1.25 \text{ x pH} - 4.0417)/4 = 0.3125*\text{pH}-1.01 \qquad (1)$$

This scalar was calculated for each grid cell and applied to the emission factors shown on Table 2 prior to calculating emissions. The EEA[10] reported that although most fertilizer displayed an increase in ammonia emissions with an increase in soil pH, this relationship is not true for urea. Therefore, soil pH scalars were not applied to urea emission factors as research has indicated that urea emissions are not affected by initial soil pH (It is thought that the hydrolysis of urea immediately increase the local soil pH, discounting the effects of initial soil pH).

<u>Natural Soils</u>

Research has shown that the soil/plant canopy can act as either a sink or source of ammonia (Roe and Mansell, 2001)[12]. The authors recommend emission factors for twelve native soil types ranging from 1.1 kg/km^2-year for pine forest to 550 kg/km^2-year for pastureland. These recommended emission factors are based on a literature review and measurements by Corsi et al.[13,14].

Potter et al.[5] estimate ammonia emissions from native soils based on several environmental variables including monthly rainfall, surface air temperature, solar radiation, soil texture, land cover type and vegetative type. The model first calculates the available mineral N substrate for ammonia emissions and then modifies this value by applying scalars for soil surface temperature, T, pH and soil moisture content, M. The scalars are of the form

$$\{1/[1 + 10^{(0.09018+2729.92/(273.16+T)-c*pH)}]\}*(1-M) \qquad (2)$$

where c is a constant which determines the sensitivity to pH. The authors used c=1.3, consistent with measurements they had made, and c=10, to produce results with minimal pH effects. Ammonia emissions are calculated for seven non-agricultural soil types.

Corsi et al.[15] (2002) note that, although their measurements in pine and oak forests yielded much lower emission factors than the averages predicted by Potter et al.[2] (see above), when corrected for the more acidic environment of the Texas forest compared to California forests, the emission factors are comparable. Battye et al.[5] provide a recommended emission factors for five non-agricultural soil types, ranging from 1.2 kg-NH_3/ha-yr or 120 kg-NH_3/km2-yr for Forests to 0.1 kg-NH_3/ha-yr, or 10 kg-NH_3/km2-yr for barren or built up land.

It is difficult to compare soil emission factors cited by various researchers because the land cover, or soil type, categories for which emission factors are reported are not consistent. The soil emission factors used for this inventory, and the source of the factors, are shown on Table 3.

Domestic Sources

Domestic sources of ammonia emissions in the current inventory include human respiration and perspiration, cloth and disposable diapers and cats and dogs. The current inventory uses emission factors for domestic sources as developed by Chitjian et al.[16] (2000). Emission factors for domestic sources are presented on Table 4.

Temporal Allocation

Livestock Operations

Ammonia emissions from livestock display both a seasonal and diurnal variation consistent, in general, with increased ammonia emissions associated with warmer temperatures. Seasonal allocation factors based on inverse modeling results have been developed by Gilliand et al.[17] (2002), and subsequently adjusted to reflect the current ORD-recommended emission factors (EPA, 2002)[6]. The seasonal allocation factors are presented in Table 5.

Aarninck[18] (1997) and Harris[19] (2001) report diurnally varying emissions at swine houses. Aarnink reported an approximately 10% increase in daytime emissions over nighttime emissions. Chinkin et al.[1] document a theoretical equation developed by Russell and Cass in 1986. This model predicts diurnal emission changes from meteorological variations (Sadeghi and Dickson, 1992)[20]. The Russell and Cass equation (Equation 3) relates hourly ammonia emission rates to temperature and wind speed as follows:

$$Ei \propto [2.36^{(Ti-273)/10}]\ Vi\ A \qquad (3)$$

where:
Ei = emission rate at hour i from animal waste decomposition
A = daily total emission rate for ammonia from animal waste = $\sum$ Ei
Ti = ambient temperature in degrees Kelvin at hour i
Vi = wind speed in meters per second (m/s) at hour i

De Visscher et al.[21] (2002) present a process-based model to predict ammonia emissions from lagoons at swine facilities. The model demonstrates good correlation to measured results, especially at wind speeds less than 15 meters/second. The model provides insight into the dependence of ammonia emission rates on temperature, pH and wind speed. Predicted ammonia emissions vary exponentially with temperature, with a marked increase at approximately 20^o C. Predicted ammonia emissions vary exponentially with pH, with a marked increase at approximately pH 8. Predicted ammonia emissions vary linearly with wind speed.

Pinder et al.[5] (2003) report model results indicating a seven-fold seasonal variation of ammonia emissions from dairies in some counties. The counties displaying the greatest seasonal variations were from the cold winter states of the northeast and northern Midwest. The authors attribute this variation to greater seasonal climate variation, winter confinement and delayed manure application. Anderson et al.[22] (2003) analyze several data sets on ammonia losses from livestock waste and demonstrate an increase in ammonia volatilization with increased temperature. The authors note the large variability in the data sets due to the numerous parameters that affect volatilization.

In the current inventory, Equation (3) is used to provide diurnal profile. This approach is consistent with first principal assumptions and with measurement showing increased ammonia release with increased temperature and wind speed. Seasonal allocation factors are based on those presented in Table 5. Although empirically based, both the seasonal and diurnal profiles are consistent with the theory that greater temperatures and greater wind speeds will result in larger ammonia volatilization rates.

Fertilizer Use

In the case of both fertilizer and livestock emissions, farm management practices, which vary geographically across the country, play an important role in determining appropriate emission factors, and can contribute to temporal variation in emissions.

Chinkin et al.[1] (2003) report that Midwest Research Institute[23] (1998) found a diurnal variation in ammonia emissions from fertilizer application that followed temperature patterns. The authors discuss the first principal model developed by Potter et al.[2] (2001) but were not able to verify the scientific integrity of the model. Roelle and Aneja[24] (2001) measured ammonia fluxes from intensively managed agricultural soils (a commercial hog operation) and determined that soil temperature plays an important role in the variability of ammonia emissions and suggests that an approach similar to the biogenic emission inventory system land use and temperature model for NO emissions may be useful in modeling biogenic ammonia emissions. The Midwest Research Institute[24] found that hourly emission rates of ammonia from fertilizer applications exhibit diurnal patterns that follow temperature patterns

In a study of ammonia in an intensively managed pasture of rye grass, van Hove et al.[25] (2001) report counteracting effects from temperature, resulting in a stomatal compensation point (emission potential) that is constant throughout the seasons. Measured stomatal compensation concentrations indicate that the grass canopy is unlikely to be a major source of ammonia emissions.

Seasonal variations in ammonia emissions from fertilizer will results from seasonal variations in fertilizer application rates. The current inventory bases seasonal profiles on the CMU monthly profile. This profile will be adjusted, if appropriate, based on more detailed fertilizer application timing information we will pursue from state agencies. Diurnal variations in fertilizer emissions are expected as temperature and wind speed affect ammonia production and volatilization. Based on generalized first principles the current inventory used the diurnal model developed by Russell and Cass presented above as Equation 3.

Natural Soils

The temporal variation of soil emissions is obtained through application of Equation 2 above for the adjustment of the emission factors described above, which are temporally resolved.

Domestic Sources

Domestics ammonia sources are assumed constant in time as no specific information on temporal variations were found in the literature reviewed for the project.

Activity Data

For livestock operations, animal headcounts were obtained from the National Agricultural Statistic Services (NASS) county livestock files (NASS, 2003)[26]. Fertilizer emissions are based on fertilizer use data. Activity data was obtained from the Association of American Plant Food Control Officers Association (AAPFCO, 2003)[27], the USDA agricultural census (2001)[28], and the county crop files (NASS, 2003)[26].

The total area covered for each land-use category from the National Land Cover Database (NLCD), described below, provides the activity data for estimating soil emissions.

Activity data for most domestic sources is total population, which is readily available (US Census Data). Pet populations are derived from total population based on ratios presented in Dickson et al.,[29] (1991), and national pet population data from Florida Cooperative Extension Service, Institute of Food and Agricultural Sciences, University of Florida.

Spatial Allocation

The spatial allocation of ammonia emissions is accomplished as part of the GIS-based model developed for the project. Population density and land use/land cover (LULC) form the basis for spatial allocation of emission sources considered in the current inventory. The LULC data used in the project are based on the National Land Cover Data developed by the USGS.

The National Land Cover Data (NLCD) was developed as part of a cooperative project between the U.S. Geological Survey and the U.S. Environmental Protection Agency to produce a consistent land cover data layer for the entire conterminous U.S. based on 30-meter Landsat thematic mapper (TM) data. The NLCD was developed from TM data acquired from the Multi-Resolution Land Characterization (MRLC) Consortium, a partnership of federal agencies that produce or use land cover data. The NLCD datasets are available as flat generic raster image files which are easily imported into a GIS (e.g., Arc/INFO) and are provided in an Albers Conic Equal Area projection at a spatial resolution of 30-meters. The data can be obtained from the following URL http://edcwww.cr.usgs.gov/pub/edcuser/vogel/states/. Figure 1 displays the NLCD for California and provides an example of the display cap[abilities of the GIS-based model developed for the project.

For livestock operations, confined animal feeding operations (CAFOs) are treated as point sources where specific CAFO locations are available. The remaining livestock ammonia emissions are allocated to grasslands and pasture lands. Fertilizer emissions are spatially allocated as function of agricultural land use. Likewise, natural soil emissions are as allocated as a function of land use. Domestic sources are spatially allocated by population.

Environmental Data

Environmental data required in the development of the ammonia emission inventory using the procedures and equations described above include meteorology and soil pH and moisture. Meteorological data, including soil moisture, is derived from MM5 model simulations for 2002 on the National RPO modeling domain. Soil pH is derived from the CONUS-SOIL dataset (Miller and White, 1998)[30].

GIS-MODEL DEVELOPMENT

ENVIRON Corporation recently developed an ammonia-capable GIS-based (ArcINFO 7.2) emission modeling system for California (ENVIRON, 2001)[31]. For many ammonia emission source categories, emission estimates and resolution are intimately linked to land cover/land use (LULC) characteristics and environmental variables.

For the WRAP NH_3 project described here, this model has been enhanced and expanded to include all states in the WRAP modeling domain. The model is developed to be efficient, robust and easily applied to any region and ammonia emission source categories for which appropriate data is available using the latest version of ESRI's GIS, ArcGIS, and the Arc Macro Language (AML). The model includes a user-friendly graphical user interface (GUI) to facilitate easy implementation. It includes the capability to easily modify activity data, emission factors and associations between ammonia emission source categories LULC characteristics. Extensive display and quality assurance features are also be incorporated into the modeling system. The model incorporates the improved estimation algorithms using environmental parameters and thus provides a user friendly streamlined process. The model has been adapted for use across the entire WRAP domain and includes the flexibility necessary to take advantage of the best available data for a given area, regardless of the source and format of the data.

The model will be populated with all the necessary data to produce an ammonia inventory for the US portion of the WRAP domain for 2002. Limitations associated with the project schedule and resources precluded the development of the necessary activity data for the portions of the WRAP domain in Canada and Mexico. The model will be used to produce a 2002 ammonia inventory for the WRAP 36 kilometer domain. Gridded ammonia emission estimates for Canada and Mexico can be obtained from existing WRAP modeling inventories.

The development of the GIS-based model is more fully documented by Chitjian and Mansell (2003)[4]

RESULTS

The year 2002 ammonia emissions estimates are currently being compiled and draft estimates should be completed in the summer of 2004. Figure 2 displays an example of gridded ammonia emissions from domestic sources. Note that the emission estimates shown in the figure are based on preliminary data and are presented to illustrate the spatial allocation and gridding capabilities of the model.

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Table 1. Comparison of Livestock Emission Factors (kg/Animal-year).

	Battye et al (2003) Recommended	**Battye et al (2003) Handbook**	**Chinkin et al (2003) Recommended**	**Doorn et al., (2002) Measured**	**Keener et al. (2001)**	**Pinder et al (2003)**	**European Environment Agency (2002)**
Dairy Cow	28	20-70	25			13.1-55	28.5
Beef Cow	10.2	9-18	9				14.3
Poultry	0.28, 0.37	0.1-0.4	0.1		0.16, 0.52		0.28, 0.37
Swine	11.4	9.5-13.5	7	7			6.39, 16.43
Horses	8		12.2				8
Sheep	1.34		3.4				1.34

Table 2. Fertilizer Emission Factors.

	Battye et al., 1994	**EEA, 2002 Group I**	**EEA, 2002 Group II**	**EEA, 2002 Group II**	**Pottet et al., 2001**
Anhydrous Ammonia	1	4	**4**	4	2.4
Aqua ammonia	1				2.4
Nitrogen solutions	2.5	8	**8**	8	2.4
Urea	15	20	**15**	15	2.4
Ammonium nitrate	2.1	3	**2**	1	2.4
Ammonium sulfate	8	15	**10**	5	2.4
Calcium ammonium nitrate		3	**2**	1	2.4
Ammonium thiosulfate	2.5				2.4
Other straight nitrogen	4				2.4
Ammonium phosphates	4.8	5	**5**	5	2.4
N-P-Ka		3	**2**	1	2.4
Potassium nitrate					2.4

Table 3. Soil Emission Factors (kg/km^2-yr).

Code	**Description**	**Emission Factor**	**Source**
11	Open Water	0	Best Estimate
12	Perennial Ice/Snow	0	Best Estimate
21	Low Intensity Residential	10	Battye et al. (2003), Built-up
22	High Intensity Residential	10	Battye et al. (2003), Built-up
23	Commercial/Industrial/ Transportation	10	Battye et al. (2003), Built-up
31	Bare Rock/Sand/Clay	10	Battye et al. (2003), Barren Lands
32	Quarries/Strip Mines/Gravel Pits	10	Battye et al. (2003), Barren Lands
33	Transitional	10	Battye et al. (2003), Barren Lands
41	Deciduous Forest	174	Average of Battye et al (2003) Forests, Chinkin et al. (2003) Oak Forest and Temperate Forest/Woodland/Shrubland
42	Evergreen Forest	54	Average of Battye et al (2003) Forests, Chinkin et al. (2003) Oak Forest and Temperate Forest
43	Mixed Forest	114	Average of Evergreen and Deciduous Forest
51	Shrubland	400	Chinkin et al. (2003) Temperate

Code	Description	Emission Factor	Source
			Forest/Woodland/Shrubland Battye et al (2003) Shrubland
61	Orchards/Vineyards/Other	0	Accounted for by fertilizer
71	Grasslands/Herbaceous	400	Chinkin et al. (2003), grasslands
81	Pasture/Hay	0	Accounted for by fertilizer
82	Row Crops	0	Accounted for by fertilizer
83	Small Grains	0	Accounted for by fertilizer
84	Fallow	205	Average of grasslands and Battye et al. (2003), Barren Lands
85	Urban/Recreational Grasses	400	Chinkin et al. (2003), grasslands
91	Woody Wetlands	400	Chinkin et al. (2003) Temperate Forest/Woodland/Shrubland Battye et al (2003) Shrubland
92	Emergent Herbaceous Wetlands	400	Chinkin et al. (2003) Temperate Forest/Woodland/Shrubland Battye et al (2003) Shrubland

Table 4. Domestic Emission Factors.

Source	Emission Factor	Unit
Cats	0.348	lb N/cat/yr
Dogs	2.17	lb N/dog/yr
Human Perspiration	0.55	lb/person/yr
Human Respiration	0.0035	lb/person/yr
Cloth Diapers	6.9	lb/infant/yr
Disposable Diapers	0.36	lb/infant/yr

Table 5. Monthly Livestock Allocation Factors.

Month	Temporal Allocation Factor
January	67
February	75
March	75
April	82
May	126
June	164
July	183
August	154
September	115
October	73
November	51
December	51

Figure 1. NLCD for California.

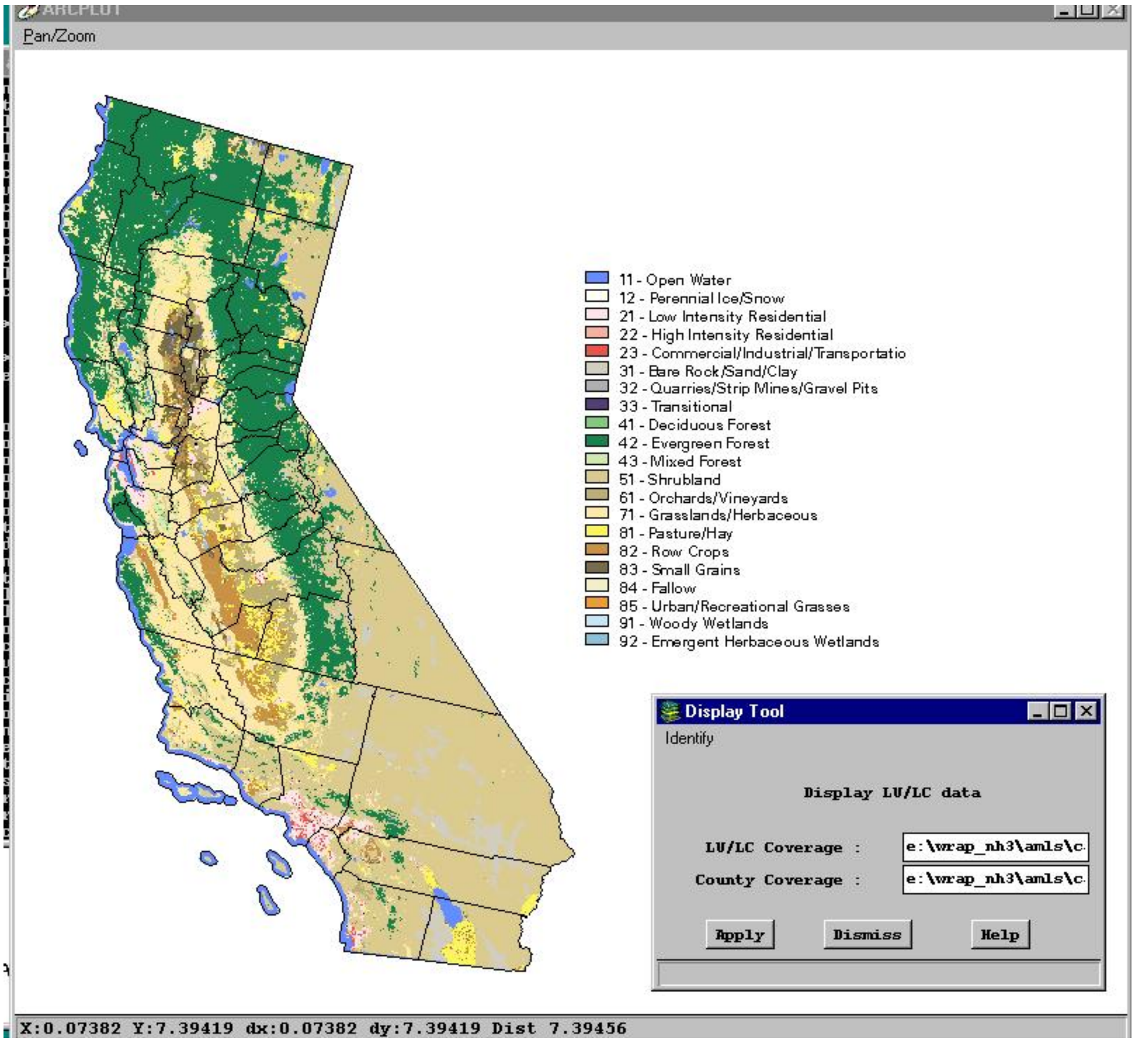
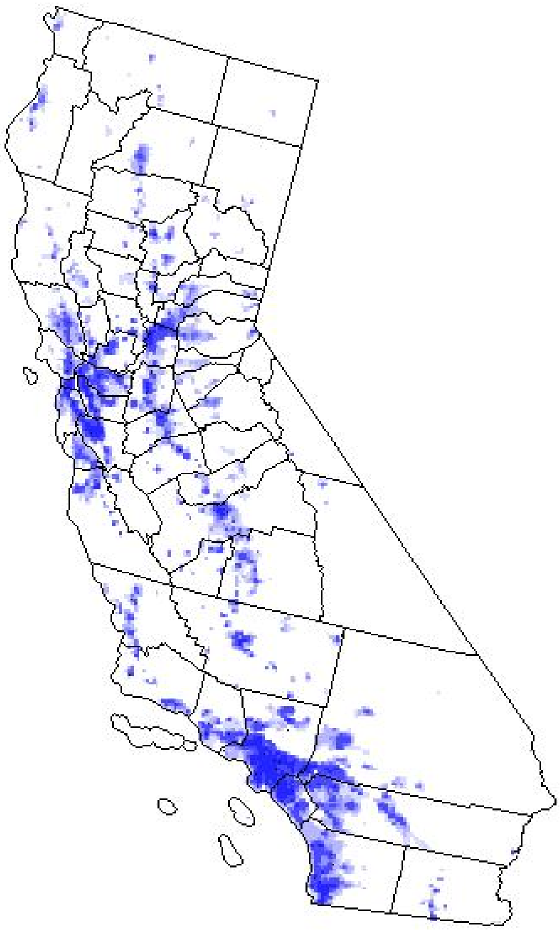


Figure 2. Example of gridded domestic ammonia emissions for California.



KEYWORDS

Ammonia
Emission Inventories
Spatial Allocation
Emissions Modeling
GIS
WRAP