An Improved Ammonia Inventory for the WRAP Domain

Literature Review

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Literature Review: WRAP Ammonia Inventory

1. Introduction.

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 emission 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. WRAP is currently providing 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. The objective of this literature review is to identify recent research in NH3 emissions and to described how it will be used in the new WRAP NH3 emissions inventory.

Two recent studies summarize much of the new research prior to 2002. A recent study by Chinkin et al. (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 and estimation methods. Potter et al. (2001) developed an inventory of fertilizer and soil ammonia emissions for the State of California using a first principal model. The Potter et al. model relies on detailed high-resolution crop coverage and farm management practices specific to each of California's growing regions, and it provides methodologies for estimating and distributing ammonia emissions from these sources based on environmental parameters.

Because Chinkin et al. (2003) provides a comprehensive review as of its publication date, we concentrated our literature search to research published since January 2002. This search resulted in the identification of the following eleven recently published works not discussed in the Chinkin et al (2003) review:

- Analysis of Atmospheric Ammonia budget for the Kanto region, Japan (Sakurai and Fujita, 2002.) Provides data on seasonal variation of ammonia emissions.
- Ammonia Emissions from Intensive Dairying: A comparison of contrasting systems in the United Kingdom and New Zealand (Jarvis and Ledgard, 2002.) Provides information relating ammonia emissions at dairy farms to farm management practices.

- The annual variation in stomatal ammonia compensation point of rye grass (*Lolium perenne L.*) leaves in an intensively managed grassland (van Hove et al., 2002) provides data relating to seasonal variation of ammonia emissions from soils and crops.
- Estimates of the emission rates from light-duty vehicles using standard chassis dynamometer test cycles (Durbin et al., 2002). Provides data on tailpipe ammonia emissions.
- Ammonia Emissions from dairy farms: Development of a farm model and estimation of emissions from the United States (Pinder et al., 2003). Provides estimates of dairy cow emission factors and provides information on seasonal variation of dairy emissions.
- Evaluation and improvement of Ammonia Emission Inventories (Battye et al., 2003) provides a current review of emission factors for soils and livestock.
- Characterization of ammonia emissions from soils in the upper coastal plain, North Carolina (Roelle and Aneja, 2001). Provides data regarding agricultural soil emissions and the dependence on environmental parameters.
- Atmospheric NH_3 concentration and N-balances for 1.6 million caged Layer facility-Manure belt/composting vs. deep pit (Keener et al., 2001). Provides data for poultry emission factors
- Development of an emission factor for ammonia emissions from US swine farms based on field tests and application of a mass balance (Doorn et al., 2002). Provides data on swine emission factors.
- Ammonia emissions from anaerobic Swine lagoons: Model development (De Visscher et al., 2002). Provides information on emission factors and the dependence of ammonia emission rates on environmental parameters
- Airborne reduced nitrogen: ammonia emissions from agriculture and other sources (Anderson et al., 2003). Provides a review of several data sets relating ammonia emissions to environmental parameters.

2. Temporal Variation.

An ammonia emission inventory is typically estimated based on emission factors for individual sources of ammonia and activity levels for that emission source. Frequently the activity data is available only for political boundaries, e.g., county or state activity, and for daily, monthly or annual activity. Because air quality models require emissions inputs for hourly resolution and spatially gridded model domains, the emission inventory must then be distributed temporally and spatially throughout the modeling domain to produce a model ready inventory.

Temporal variation in ammonia emissions from a given source can result from either a variation in activity levels of that source (i.e., fertilization schedules, traffic patterns) or a variation in the parameters influencing ammonia release (i.e., wind, temperature.)

Sakurai and Fujita (2002) concluded that warm season (April to September) ammonia emission rates for the Kanto region comprise 70% of the total annual emissions and cold season (October to March) ammonia emissions are 30% of the annual total. The authors also indicate that the seasonal variation is larger in more rural areas than urban areas. The seasonal variation in ammonia emissions is attributed to seasonally varying temperatures. This study considered all emission sources including agricultural, urban and natural.

The National Academy of Science (2002) note that animal emission factors are not well characterized and recommend a process-based modeling approach to estimate emissions from concentrated feeding operations

Livestock Emission Factors

Chinkin et al. recommend the use of the Office of Research and Development (ORD) cattle, swine and poultry emission factors (EPA, 2002). A limited summary (not all animal categories are included) of livestock emission factors recommended by Chinkin et al. and other researchers described below is shown on Table 1.

Battye et al. (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 an interesting comparison of these factors to estimates determined from the Midwest Plan Service, a handbook that provides estimated 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 the ORD factors recommended by Chinkin et al. (2003). The poultry emission factors recommended by Battye et al. (2003), 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. (2003), 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 (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. These factors are consistent with the ORD recommended emission factor for dairy cows.

Keener et al. (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. The authors state that results based on airflows and concentrations are not as certain, as airflows did not remain constant. July testing resulted in similar emission factors of 0.458 kg-NH_3/animal-yr for the deep pit system and 0.165 kg-NH_3/animal-yr for the belt/compost system. These results do not indicate a trend towards higher summertime emissions at might be expected. These emission factors are fairly consistent with those recommended by Battye et al. (2003) but are much greater than those recommended by ORD.

Doorn et al. (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. (2002) also provides a comparison to several other researchers’ results in addition to those reviewed by Battye et al. (2003). The authors noted that testing showed a significant diurnal cycle but provided no quantification of the variation in emission rates. The authors did note that daytime emissions represent an “upper bound” because of increased temperature and increased animal activity during daytime hours. This emission factor is consistent with the ORD factors and the Battye recommended factors

Jarvis and Ledgard (2002) compare two significantly different dairy farms in the U.K. and New Zealand. The authors find a more than two-fold increase in emissions at the U.K. farm compared to the New Zealand farm. The authors attribute this difference to the need for winter housing in U.K., almost all of the difference is associated with the storage and application of slurry resulting from winter housing.

3. Livestock Temporal.

Chinkin et al cite a lack in consistency of results quantifying temporal variations in ammonia emissions from livestock. However, a preponderance of the studies cited concluded ammonia emissions from livestock display both a seasonal and diurnal variation consistent, in general, with increased ammonia emissions associated with warmer temperatures.

Chinkin et al recommend seasonal allocation factors based on those proposed by Gilliand et al. (2002), which are based on inverse modeling results. The factors were adjusted to reflect the current ORD-recommended emission factors (EPA, 2002), which were not available at the time the modeling was performed by Gilliand et al. The adjusted factors are shown on Table 2.

Inspection of Table 2 indicates a 3-4-fold increase in emissions during the warmest months and minimum emissions during the late fall, as opposed to the coldest months. The minimum in fall is explained by the relatively dry conditions at this time of the year.

Chinkin et al report that both Aarninck (1997) and Harris (2001) report diurnally varying emissions at swine houses. Aarnink (1997) reported an approximately 10% increase in daytime emissions over nighttime emissions. The authors recommend the diurnal profile, shown on Table 3 and Figure 1, based on this information. Citing a lack of quantitative data to support a diurnal profile for other livestock categories, and the expectation that other categories should also display a diurnal variation similar to swine houses, Chinkin et al recommend use of this profile for all livestock categories.

Both the seasonal and diurnal profiles recommended, although empirically based, are consistent with the theory that greater temperatures and greater wind speeds will result in larger ammonia volatilization rates.

De Visscher et al. (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. (2003) report 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. (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 which affect volatilization: they note for instance, an unexpectedly high volatilization rate at near freezing temperatures which they suggest is a result of frozen soil blocking the infiltration of slurry, eventually leading to increased volatilization.

4. Fertilizer Emission Factors.

The model developed by Potter et al. (2001) estimates ammonia emissions from fertilizers as a function of fertilizer management practices and several environmental variables. The authors noted that results from emissions experiments indicate changes in pH (>7.5) had the most significant effect on ammonia emissions (Dewes, 1996). 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. There is no distinction made between different types of fertilizers.

5. Fertilizer Temporal.

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. (2003) report that Midwest Research Institute (1998) found a diurnal variation in ammonia emissions from fertilizer application that followed temperature patterns. The authors recommend a diurnal profile of nitrous oxide emissions, as shown on Table 3 and Figure 1, measured by Anderson and Levine (1987). The authors discuss the first principal model developed by Potter et al. (2001) but were not able to verify the scientific integrity of the model.

Roelle and Aneja (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 (Log_{10} NH_3-N flux = .054T_{soil}+0.66; R^2=0.71) 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.

In a study of ammonia in an intensively managed pasture of rye grass, van Hove et al. (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.

6. Soils.

Chinkin et al. warn that research has shown that the soil/plant canopy can act as either a sink or source of ammonia (Roe and Mansell, 2001). 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. (2000, 2002a.) Based on the large degree of uncertainty, the authors recommend that soils emissions not be included in the ammonia inventory.

Potter et al 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 $\{1/[1 + 10^{(0.09018+2729.92/(273.16+T)-c*pH)}]\}*(1-M)$, 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 soils types. Emission factors derived from these values range from 6.5 kg/km^2-year, for evergreen needle leaf forests, using a moderate pH effect to 206 kg/km^2-year for mixed forests, using the minimal pH effect.

Corsri et al. (2002b) note that, although their measurements in pine and oak forests yielded much lower emission factors than the averages predicted by Potter et al. (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. (2003) 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.

7. Mobile Sources.

Durbin et al. 2002 report ammonia emissions of 54 mg ml-1 for a fleet 39 in-use light duty vehicles over the federal test procedure (FTP) driving cycle. Although this fleet included a wide range of years and emission control technologies, it may not accurately reflect the national fleet.

Table 1. Comparison of Livestock Emission Factors.

	Dairy Cow	Poultry	Swine
Battye et al (2003) Recommended	28	0.28, 0.37	6.4, 16.4
Battye et al (2003) Handbook	20-70	0.1-0.4	5-17
Chinkin et al (2003) Recommended	25	0.1	6.8
Doorn et al., (2002) Measured			7
Keener et al. (2001)		0.16, 0.52	
Pinder et al (2003)	13.1-55		

Table 2. Monthly Livestock Allocation Factors, Chinkin et al.

Month	Factor (% annual average)
January	67
February	75
March	75
April	82
May	126
June	164
July	183
August	154
September	115
October	73
November	51
December	51

Table 3. Diurnal Livestock Emission Profile, Chinkin et al.

Hour of Day	Livestock	Fertilizer
12:00 AM	3.9	2
1:00 AM	4.0	2
2:00 AM	4.0	2
3:00 AM	4.1	2
4:00 AM	4.1	2
5:00 AM	4.2	2.1
6:00 AM	4.2	2.8
7:00 AM	4.2	4.1
8:00 AM	4.2	7
9:00 AM	4.3	7.4
10:00 AM	4.3	8.2
11:00 AM	4.3	8.2
12:00 PM	4.3	8.1
1:00 PM	4.3	7.8
2:00 PM	4.3	6.5

3:00 PM	4.3	4.1
4:00 PM	4.2	4.1
5:00 PM	4.2	3.1
6:00 PM	4.2	2.9
7:00 PM	4.2	2.9
8:00 PM	4.1	2.9
9:00 PM	4.1	2.9
10:00 PM	4.0	2.9
11:00 PM	4.0	2

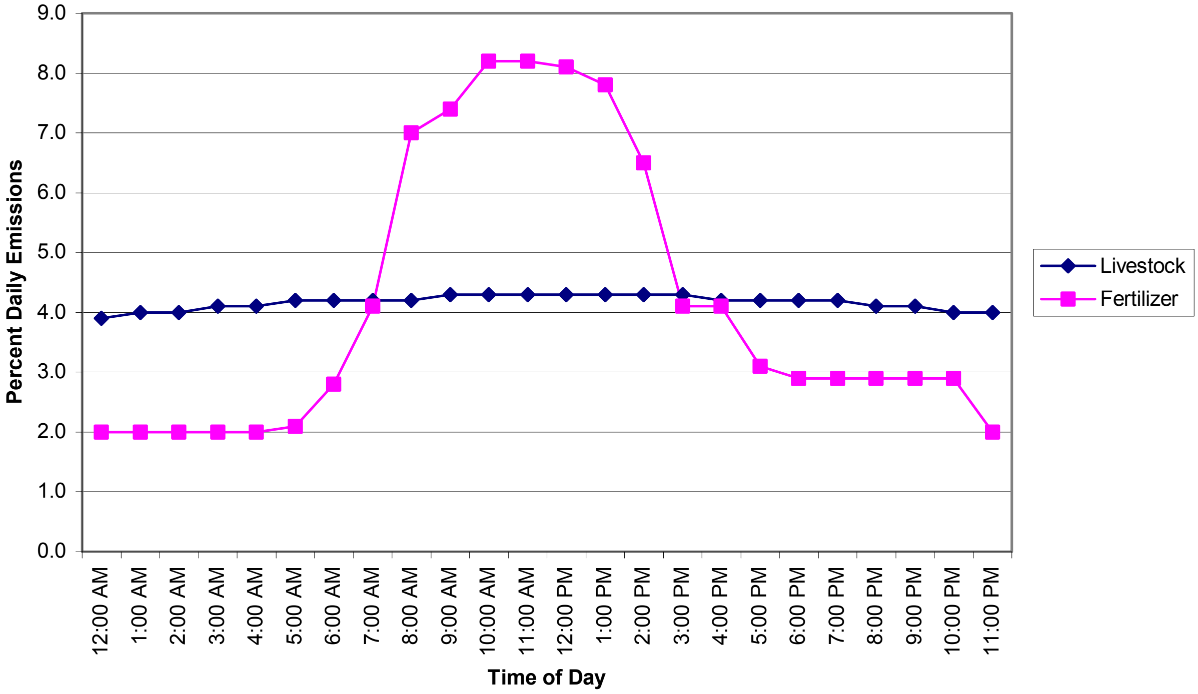

Figure 1. Livestock and Fertilizer Diurnal Emission Profile, Chinkin et al.

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