

Ammonia Assessment from Agriculture: U.S. Status and Needs

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Recent studies suggest that human activities accelerate the production of reactive nitrogen on a global scale. Increased nitrogen emissions may lead to environmental impacts including photochemical air pollution, reduced visibility, changes in biodiversity, and stratospheric ozone depletion. In the last 50 yr, emissions of ammonia (NH_3), which is the most abundant form of reduced reactive nitrogen in the atmosphere, have significantly increased as a result of intensive agricultural management and greater livestock production in many developed countries. These agricultural production practices are increasingly subject to governmental regulations intended to protect air resources. It is therefore important that an accurate and robust agricultural emission factors database exist to provide valid scientific support of these regulations. This paper highlights some of the recent work that was presented at the 2006 Workshop on Agricultural Air Quality in Washington, D.C. regarding NH_3 emissions estimates and emission factors from agricultural sources in the U.S. and Europe. In addition, several best management practices are explored as the scientific community attempts to maximize the beneficial use of reactive nitrogen while simultaneously minimizing negative environmental impacts.

BIOLOGICALLY active, photochemically reactive, and radiatively active nitrogen compounds in the atmosphere, hydrosphere, and biosphere are collectively referred to as reactive nitrogen (Galloway et al., 2003). Over the past few decades, human activities leading to the production of reactive nitrogen from diatomic nitrogen (N_2) exceed that of nitrogen fixation in the natural terrestrial ecosystem at the global scale (Galloway et al., 2004).

Ammonia (NH_3) is the most reduced form of reactive nitrogen. It is also the most abundant alkaline constituent in the atmosphere (Aneja et al., 2006a). Figure 1 illustrates the major processes (emissions, chemical transformation, transport, and removal) that drive the global cycle of NH_3 in the atmosphere (Aneja et al., 2006b,c). In the past 50 yr, emissions and subsequent deposition of NH_3 have increased significantly in parallel with the development of intensive agricultural management and increased livestock numbers (Sutton et al., 1993). Globally, domestic animals are the largest source [32×10^{12} g $\text{NH}_3\text{-N}$ (ammonia-nitrogen) yr^{-1}] of atmospheric NH_3 , comprising approximately 40% of natural and anthropogenic emissions combined (Schlesinger and Hartley, 1992). Synthetic fertilizers and agricultural crops together contribute 9×10^{12} g $\text{NH}_3\text{-N}$ yr^{-1} (12% of total emissions) (Schlesinger and Hartley, 1992). The first Workshop on Agricultural Air Quality: State of the Science (Aneja et al., 2006a) was structured to help scientists, industry, policymakers, and regulators make optimal choices about issues confronting agricultural practices to maximize the benefits and reduce the detrimental environmental effects of current food, fiber, and feed production activities. This paper focuses on issues surrounding ammonia emissions, its transport, transformation, and fate.

Ammonia Emissions

Emissions of air pollutants, particularly ammonia, during agricultural operations are an important emerging research area in the U.S., best studied with interdisciplinary approaches that can inform policymakers of the costs and benefits of various mitigation options. Data on agricultural emissions of regulated pollutants, nuisance odors, and fugitive dust often either do not exist or are insufficient to develop appropriate policy nationally. Emissions estimates from

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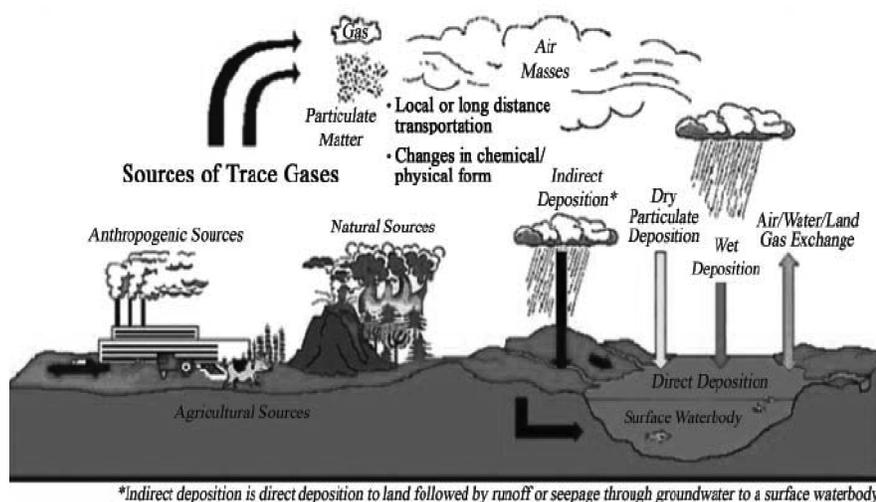


Fig. 1. Atmospheric emissions, transport, transformation, and deposition of trace gases (Source: Aneja et al., 2003, 2006b).

emerging agricultural producers in Southeast Asia (e.g., China and India) and other developing countries are even more limited. Table 1 shows the total 2002 estimated NH₃ emissions from various animal husbandry operations in the United States (USEPA, 2005). Table 2 lists ammonia emission factors reported for the Czech Republic, Denmark, and the Netherlands. Several other European countries have also developed specific NH₃ emission factors for intensive animal agriculture practices. Some countries (e.g., the U.S.) are currently in the process of developing national emission factors for both animal and crop agriculture (e.g., Table 3). Unfortunately, the process of developing appropriate emission factors is hindered in the U.S., because relatively few experiments

Table 1. Ammonia emission estimates in the United States for the year 2002.†

Source	Ammonia emissions	
	(tons/yr)	% of Total emissions
Animal		
Dairy(dairy cows and dairy heifers)	558,094	12.71
Beef(beef cattle, bulls, and calves)	656,648	14.95
Poultry(chickens and turkeys)	664,238	15.12
Swine(breeding and marketing pigs)	429,468	9.78
Sheep	24,835	0.57
Goats(milking and Angora goats)	14,028	0.32
Horses	71,285	1.62
Total animal	2,418,595	55.07
Fertilizer	1,140,396	25.97
TOTAL AGRICULTURE	3,558,991	81.03
Other		
Chemical and applied product MFG	23,123	0.53
Fuel comb. elec. util.	30,256	0.69
Fuel comb. industrial	15,959	0.36
Miscellaneous	282,166	6.42
Fuel comb. other	17,602	0.40
Mobile sources	289,871	6.60
Waste disposal and recycling	25,770	0.59
Other industrial processes	148,288	3.38
Total other	833,035	18.97
TOTAL EMISSIONS	4,392,026	100.0

† Source: USEPA (2005).

with adequate spatial and temporal resolution have been conducted to estimate emissions from animal production, especially concerning dairy, beef cattle, and poultry. Additionally, relevant studies often neglect to report specific information (e.g., average animal weight, total area) necessary to develop emission factors expressed as [g NH₃-N kg⁻¹ d⁻¹] in accordance with the United States Department of Agriculture (USDA) recommendations (Flesch et al., 2007; Cassel et al., 2005; Gay et al., 2003).

Developing accurate emissions inventories is also difficult because many factors (e.g., seasonality, time of day, temperature, humidity, wind speed, solar intensity, and other weather conditions, ventilation rates, housing type, manure properties or characteristics, and animal species, stocking density, and age) are involved in the generation and dispersion of ammonia. Emission factors therefore vary both spatially and temporally, and all of the parameters discussed above must be considered when used in air quality modeling. Information on ammonia emissions from fertilizer applications and confined animal feeding operations (CAFOs) provide only annual averages and often disregard spatial and seasonal variations. Furthermore, the uncertainties associated with these estimates are large and emissions estimates applied for one set of conditions or for one type of crop and animal feeding operation may not translate readily to others. In general, agricultural emission factors from European conditions have been applied for estimating ammonia emissions in the U.S. To develop effective emission control strategies in the U.S., it is imperative that appropriate U.S.-based emission factors for both gases and particulate matter (PM) be developed.

Ammonia Transformation and Fate

Once released into the atmosphere, NH₃ has a relatively short residence time of about 1 to 5 d (Warneck, 2000). When airborne, it is either readily converted to aerosol or it is subjected to dry or wet deposition processes. Ammonia is reactive with a variety of acidic atmospheric species, including nitric acid (HNO₃), hydrochloric acid (HCl), and sulfuric acid (H₂SO₄), which result in the formation of ammonium aerosols, i.e., fine particulate matter (aerodynamic diameter <2.5 μm). Due to the extended lifetime of these aerosols (about 1–15 d), nitrogen may be transported to previously pristine regions far from the pollutant sources (Aneja et al., 2001). Assuming an atmospheric residence time of 6 d and a wind velocity of 5 m s⁻¹, Irwin and Williams (1988) estimate that ammonium aerosols might travel as far as 2500 km; however, dry and wet deposition may reduce this transport significantly (Erisman and Draaijers, 1995).

Ammonia Transformation and Fate

Both ammonia and subsequently derived ammonium (NH₄⁺) may be removed from the atmosphere through both wet and dry deposition. Dry deposition occurs by diffusion of NH₃ in the atmospheric boundary layer and the surface layer (Niyogi et al.,

2003). Wet deposition occurs by below cloud scavenging (washout) and rainout (in-cloud processes). Fine particulate NH_4^+ is efficiently removed from the atmosphere mainly through wet deposition. Overall, wet deposition is more important in regions with low NH_3 emissions. Conversely, dry deposition is more important in regions of high NH_3 emissions (Erisman and Draaijers, 1995; Krupa, 2003; Niyogi et al., 2006).

Scientific information suggests that reactive nitrogen (e.g., NH_3) is accumulating in the environment, and that nitrogen cycling through biogeochemical pathways has a variety of environmental consequences including acidification and eutrophication, photochemical air pollution, reduced visibility, ecosystem fertilization, global warming, and stratospheric ozone depletion (Galloway et al., 2003). Lehmann et al. (2005) have reported statistically significant increases in wet deposition of ammonium and dissolved inorganic nitrogen in much of the United States over an eighteen year period from 1985 to 2002. A number of studies have evaluated the effects of nitrogen deposition (Paerl, 1995, 1997; Paerl and Whitall, 1999; Spiek et al., 1990; Sørensen et al., 2003). Significant excess nitrogen deposition has occurred in the eastern coastal areas of the United States (Paerl, 1995). A particular area of concern is the coastal rivers and their estuaries. Atmospheric deposition of nitrogen compounds may contribute as much as 35 to 60% of total nitrogen loading to North Carolina coastal waters (Paerl, 1995). This excess nitrogen can result in toxic and non-toxic phytoplankton blooms, which can lead to fish kills and reductions of 'clean water' species (Paerl, 1995). In cooler climates of Europe (e.g., Denmark) it is unlikely that atmospheric deposition events trigger algae blooms directly (Hasager et al., 2003) within the coastal waters surrounding Denmark. However, the deposition amounts are high enough to increase chlorophyll production by 20% or more in these areas. Furthermore, the atmospheric deposition constitutes a large part of the overall load in these waters and is therefore an important source for fixed nitrogen (i.e., nitrogen taken from its relatively inert molecular form (N_2) in the atmosphere and converted into nitrogen compounds such as NH_3) (Spokes et al., 2006). Soil acidification is another problem experienced in

Table 2. Ammonia emission factors for individual animals categories for some countries in Europe

Animal category	Czech Republic 2002†	Denmark 2005‡	The Netherlands 2006§
	Emission factor		Emission factor
	kg NH_3 animal ⁻¹ yr ⁻¹		kg NH_3 animal place ⁻¹ yr ⁻¹
Dairy cows	27.9	26.92	
Grazing			9.5
100% housed			11
Cows	16.2		
Beef cattle 6–24 mo			7.2
Heifers	16.2		
Heifers 6 mo-calving		5.49	
Heifer calves < 6 mo		4.00	
Calves	16.2		2.5
Bull calves 6–14 mo		8.45	
Bull calves < 6 mo		3.51	
Bulls	16.2		9.5
Other cattle	16.2	9.00	5.5
Farrowing sows (incl. piglets)			8.3
Dry and pregnant sows			4.2
Boars > 7 mo			5.5
Total Sows	17.44	7.60	
Sucking-pigs	6.5	0.18	
Large pen			0.75
Small pen			0.60
Pigs	8.3	1.01	
Large pen			3.5
Small pen			2.5
Broiler breeders < 19 wk			0.250
Broiler breeders			0.580
Broilers	0.21	26.60	0.080
Layers	0.27	36.95	
Layers < 18 wk (deep pit)			0.170
Layers + layer breeders(deep pit)			0.315
Turkey cocks and hens	0.92	29.00	0.680
Other poultry	0.21		
Geese and ducks	0.73	7.54	
Ducks; outside keeping			0.019
Ducks; inside keeping			0.210
Horses	8	8.32	
Sheep	1.34	2.41	
Goats	1.34	2.33	

† Source: Zapletal and Chroust, 2006.

‡ Source: Gyldenkaerne, personal communication, 2007.

§ Source: Starmans and Van der Hoeek, 2007.

Northern and Central Europe, due to the high density of animal operations. Van Breeman et al. (1982) identified the deposition of ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$) as the main cause of soil acidification in the Netherlands. Research conducted by Barthelmie and Pryor (1998) in the Lower Fraser Valley, British Columbia, Canada showed that NH_3 and NH_4^+ species and emissions play a particularly critical role in visibility degradation. Fine particulate aerosols have also been linked to human respiratory health problems. Studies suggest that the smaller the particle the greater the potential health effect. For example, Lippmann (1998) found fine particles ($\text{PM}_{2.5}$) to be more toxic than coarse particles (PM_{10} – $\text{PM}_{2.5}$). Donaldson and MacNee (1998) examined ultra-fine particles (<100 nm) and found that toxicity increases as particle size decreases.

On the positive side, reactive nitrogen has contributed greatly to the increase in food production, which becomes more important as the human population is expected to grow to 8 to 12 billion by the

Table 3. Ammonia emission factors from animal and crop agriculture in the US

Animal	Animal agriculture emission factor (kg-NH ₃ animal ⁻¹ yr ⁻¹)		
	USEPA NEI 2002†	Battye et al., 2003‡	USEPA Battye et al. 1994§
Dairy cow	38.1	28	40
Beef cow		10.2	27 (steers)
On feed	20.8		
Grazing	4.3		
Pigs	6.5		
Sow		16.4	16
Finishing pig		6.4	7
Poultry	.27		
Laying hen		0.31	0.31
Broiler		0.28	0.17
Sheep	3.4	1.34	3.4
Horses	12	8.0	12
Crop agriculture emission factor (kg NH ₃ /Mg N)			
Fertilizer		Aneja et al. 2003¶	USEPA; Battye et al. 1994§
N-P-K		48	48
Nitrogen solutions		30	30
Ammonium phosphates		48	48
Anhydrous NH ₃		12	12
Urea		182	182
Ammonium nitrate		25	25
Other straight nitrogen		30	30
Ammonium sulfate		97	97
Aqua NH ₃		12	12
Ammonium thiosulfate		30	30

† Source: USEPA, 2004.

‡ Source: Battye et al., 2003.

§ Source: Battye et al., 1994.

¶ Source: Aneja et al., 2003.

end of the 21st century. Therefore, the challenge for the scientific community is to find ways to maximize beneficial use of reactive nitrogen while simultaneously minimizing adverse environmental impacts. One way to approach this challenge is through the deliberate integration of reactive nitrogen research, management, and control strategies. Integrated research and control strategies that consider urban-rural air quality connections and interactions are necessary for optimal ammonia/nitrogen management.

Best Management Practices

Assessments of best management practices (BMPs) that can positively affect agricultural air quality and also provide economic feasibility are still being examined. From the papers presented at the Workshop and our review, a number of studies suggest the best options to reduce ammonia emissions could be to reduce the formation of ammonia and to increase nitrogen use efficiency. An example of a relatively simple solution to reduce ammonia production is the reduction of crude protein in animal diets. Multiple studies have found that reducing crude protein results in lower NH₃ concentrations (Frank and Swensson, 2002; Powers et al., 2006, 2007). However, crude protein is also an important component of animal diets and it remains to be studied how it may impact animal well being. In addition, when condensed tannin is added to the drinking water of both cattle and sheep, less ammonia is volatilized. The amount of nitrogen in solid and liquid waste was similar to that of regular water, but the nitrogen was nitrified/

denitrified rather than volatilized into ammonia (Kronberg, 2006). However, if this is not possible in current facilities, several BMPs and/or emission reduction options to curtail ammonia emissions from agricultural sources have been tested, mainly dealing with emissions from cattle and swine. A relatively simple solution was undertaken by Lefcourt and Meisinger (2001), who tested the addition of alum and zeolite to cattle slurry in an effort to curb the volatilization of ammonia. When alum was added at 2.5 and 6.25%, there were reductions of 58 ± 6% and 57 ± 10%, respectively. Slightly lower reductions were seen with the addition of zeolite, with additions of 2.5 and 6.25% resulting in reductions of 22 ± 6 and 47 ± 10%. Similar tests were performed by Berg (2006), with attempts to lower ammonia emissions from cattle slurry by acidifying it with lactic and nitric acid. Lactic acid was applied to reach pH levels of 5.73, 5.14, and 4.18, yielding decreased emissions of 65, 72, and 88%, respectively. Nitric acid reduced emissions by about half of the success seen with lactic acid. When nitric acid was added to reach pH levels of 5.20 and 4.49, emissions were lowered by 29 and 49%, respectively. A side effect is that although ammonia emissions are reduced, the total reactive nitrogen production increases. The costs of the additive (such tannin, alum, and zeolite) would depend on the scale of operation and is not considered in the reported studies.

More approaches to reduce dairy cattle emissions include changes in floor design and ventilation. It was found that if a flat floor in a cattle barn was scraped 96 times per day there was a 5% reduction in the ammonia emitted from the barn. Yet, with a sloped floor, reductions were much greater. With 12 scrapings per day, there was a 21% reduction and, with 96 scrapings per day, there was a 26% reduction (Bramm et al., 1997). The study does not report on the impact on other waste-handling emissions (e.g., lagoons).

Using a filtered, custom built, double-polytube ventilation system in their calf barns, Hillman et al. (1992) were able to reduce the ammonia concentration from about 5.7 mg m⁻³ in the air to about 2.9 mg m⁻³ and below; however, the impact on the total emissions remains to be studied.

Both simple and more detailed BMPs have been tested for the reduction of ammonia from swine sources. A simple solution was the addition of the manure additive Alliance to swine manure, resulting in a 24% reduction in ammonia emissions (Heber et al., 2000), while an example of a more complicated solution involves ventilation and indoor air climate control (Hartung, 2006). There was a reduction of 10 to 14% of ammonia when there was a reduction of indoor air temperature and ventilation rate. Yet, to obtain these results an evaporative indoor air cooling system with an “optimization of the fogging control with regard to a continuously complete evaporation of water” was needed. Other solutions are more complicated in design and setup, but are easier to use. Using biotrickling filters for the manure, Hansen and

Jensen (2006) were able to reduce both odor and ammonia emitted from manure. Loyon et al. (2006) found that with the use of a storage spreading system with biological treatment of manure, there was a 30 to 50% reduction with separated manure and a 68% reduction with unseparated manure.

Emission controls of ammonia down to the farm level and the optimization of agricultural production methods have been evaluated in Denmark using modeling frameworks (Ambelas Skjøth et al., 2006). These emission models are being used for explaining variations and trends in ambient ammonia concentrations as a result of changing agricultural practices (BMPs) brought about by national regulations and policies.

Promising results have been reported for reducing ammonia from swine manure through the use of an “engineered system,” i.e., a treatment plant with solid-liquid separation. Szögi and Vanotti (2006) reported a 73% reduction in ammonia emissions from the implementation of such a system. Vanotti (2006) found that when manure from such a system was applied, there was a 98.8% reduction in greenhouse gas emissions, as well as a potential for additional income of \$9,100 to \$27,500/yr (approximately \$0.91/finished pig) from implementing cleaner technology through the Supersoil program. In addition, when organic fertilizers with gypsum are applied, they can reduce ammonia volatilization by 11% (Model, 2006).

Conclusions

In the U.S., there are currently no regulations or incentive programs that require reductions in ammonia emissions. It is striking when comparing the different pollutants, sulfur dioxide (SO₂), oxides of nitrogen (NO_x), volatile organic compounds (VOCs), and ammonia (NH₃), that the extensive control measures applied to SO₂, NO_x, and anthropogenic VOCs have not been extended to ammonia, which has similar or higher contributions to e.g., PM exposure, visibility, or acidification/eutrophication. However, future policies and control measures are necessary to successfully decrease ammonia emissions and its related problems. Current research problems related to ammonia include the quantification of agricultural point and nonpoint sources and its temporal variation; re-emission in high emission/deposition areas; the atmosphere-biosphere exchange of ammonia and its effect on SO₂ deposition; the quantification of landscape processes; low-level dispersion processes; the primary and secondary emissions of PM; and the gas-to-particle conversion to PM fine.

In the past, policies to control criteria pollutants mainly focused on single pollutants, while addressing, in general, single effects. Today, multi-pollutant/multi-effect approaches are being considered e.g., in Europe, the Gothenburg Protocol under the United Nations Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air Pollution, and the EU National Emission Ceiling Directive, thus offering unique opportunities for the development of ammonia abatement measures using integrated approach strategies across multiple media.

Generally, limited data exist for estimating agricultural emissions of air pollutants (e.g., NH₃, H₂S) and public nuisances (e.g., odors, fugitive dust). Credible estimates of air emissions from

CAFOs are also complicated by factors that affect the amounts and dispersion of emissions in the atmosphere. Emissions that occur throughout the food production system must be quantified accurately to address critical air quality issues (Aneja et al., 2006c).

The papers that are part of this special section address emissions and fate of both gases (e.g., greenhouse gases, VOCs, etc.) and particulate matter. New technologies for measurements and analysis of trace gases, and transport and transformation modeling of these pollutants are explored. The BMPs being studied for gas and odor reductions and agricultural air quality, in general, appear to be in their infancy and more comprehensive studies are needed that can develop large scale, economically viable practices.

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