PERGAMON

Atmospheric Environment 37 (2003) 3873–3883

ATMOSPHERIC ENVIRONMENT

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Evaluation and improvement of ammonia emissions inventories

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Received 2 September 2002; accepted 30 April 2003

Abstract

Two case studies are performed to improve ammonia emissions inputs used to model fine particulate matter ($PM_{2.5}$ is the portion of particulate matter smaller than 2.5 μm aerodynamic diameter) formation of ammonium sulfate and ammonium nitrate. Ammonia emissions are analyzed in detail for North Carolina and the San Joaquin Valley (SJV) of California, with a focus on the Charlotte, NC, and Fresno, California metropolitan areas. A new gridded ammonia emissions inventories suitable for atmospheric modeling for the two case study cities was also developed.

Agricultural sources accounted for the bulk of ammonia emissions in both case studies. Livestock waste contributed about 80% in North Carolina and 64% in the SJV, while fertilizer application contributed about 6–7% in both domains. Forests and non-agricultural vegetation contributed 5% in North Carolina and 12% in the SJV. Motor vehicles accounted for about 6% of ammonia emissions in North Carolina and 14% in the SJV. In the Charlotte and Fresno urban areas, the distribution of emissions is less heavily weighted toward agricultural sources and more heavily weighted toward highway vehicles (highway vehicles account for an estimated 64% of emissions in Charlotte and 51% of emissions in Fresno). The emissions estimates for agricultural sources (livestock and fertilizer application) decline to approximately 14% in the winter for both the Charlotte and Fresno urban areas. Emissions estimates for soils and vegetation also decline to approximately 0 during the winter for both the Fresno and Charlotte area. As a result, motor vehicles account for a larger fraction (approximately 73% and 70% for Charlotte and Fresno, respectively) of winter ammonia emissions, particularly in the Charlotte urban area.

Keywords: Ammonia; Emissions; Inventory; Agriculture; Particulate matter

1. Introduction

Ammonium nitrate (NH_4NO_3) and ammonium sulfates (NH_4HSO_4 and $[NH_4]_2SO_4$) are important constituents of airborne fine particulate matter ($PM_{2.5}$), and can contribute significantly to visibility impairment and regional haze. These compounds are secondary particulates, formed from gaseous emissions of ammonia (NH_3), sulfur dioxide (SO_2), and nitrogen oxides (NO_x). A number of efforts are underway to model the formation of secondary particulates in the atmosphere. These models rely on detailed inventories of precursor emissions, but inventory development for ammonia has lagged behind these other precursor pollutants.

There is currently no broad air pollution control program for ammonia comparable to the programs for SO_2 and NO_x. However, the development of cost-effective control strategies for sulfates and nitrates will hinge on a thorough understanding of the relative abundance and distribution of all precursor emissions—NH_3, SO_2, and NO_x. The interrelationship among ammonia, sulfate, and nitrate might cause nitrate levels to increase in some regions as sulfates decline. On the

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1352-2310/03/$ - see front matter
doi:10.1016/S1352-2310(03)00343-1

other hand, if the concentration of ammonia exceeds the combined concentrations of sulfate and HNO_3, then reductions in sulfate would not cause offsetting increases in nitrate.

Most ammonia emissions emanate from livestock wastes (Battye et al., 1994; Aneja et al., 1998, 2000, 2001, 2003). However, forests and other plants have been shown to emit ammonia to the air. In addition, studies have shown an equilibrium between ammonia in the air and ammonium compounds in plants' leaves. As a result, trees, crops and other plants might release more ammonia if emissions from other sources are reduced. Nonetheless, some reductions in ammonia emissions are likely to occur as a result of measures designed to reduce the runoff of ammonia and other nitrogen compounds from farmlands.

While SO_2 and NO_x have been the subject of emissions inventory efforts since the 1970s, inventory development for ammonia has lagged behind these other precursor pollutants. The spatial distribution and seasonal variations of ammonia emissions are as important as the overall magnitude of emissions. Ammonium sulfate and nitrate formations are both subject to seasonal influences. A thorough understanding of the spatial distribution of emissions is critical in order to model the interactions among ammonia, SO_2, and NO_x.

The purpose of the current research was to improve ammonia emissions inputs through a case study approach. Ammonia emissions are analyzed in detail for two cities. One of these case study cities is in the western US, where NH_4NO_3 is the dominant form of secondary $PM_{2.5}$. The other is in the eastern US, where ammonium sulfates are dominant. Charlotte, North Carolina was selected as representative of an eastern city, where ammonium sulfate dominates secondary particulate matter. Fresno, in California's San Joaquin Valley (SJV), was selected as representative of a western city, where NH_4NO_3 dominates secondary particulate matter. The primary purpose of the emissions inventory improvements is to support atmospheric models for secondary particulate formation. However, the emissions inventory improvements can also be used in modeling deposition to surface waters.

Existing inventories for the two case study areas were reviewed, including: a statewide North Carolina ammonia inventory prepared by the North Carolina Department of Environment and Natural Resources (NC DENR), the SJV ammonia inventory sponsored by the California Air Resources Board (CARB), and ammonia emissions extracted from the EPA's National Emissions Trends (NET) inventory (NCDEHNR, 1997; Coe et al., 1998; USEPA, 2000a). A new gridded ammonia emissions inventories suitable for atmospheric modeling for the two case study cities was then developed. In addition, to facilitate comparison with previous emissions inventories, county-level inventories for the state of North Carolina and the SJV region of California were prepared. Figs. 1 and 2 show the gridded inventory domains for the Charlotte and Fresno, as well as the domain for California's existing SJV inventory. The base year for our analysis was 1997.

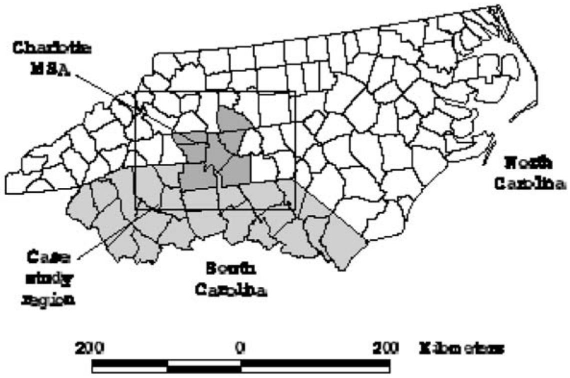

Fig. 1. Charlotte area inventory domain.

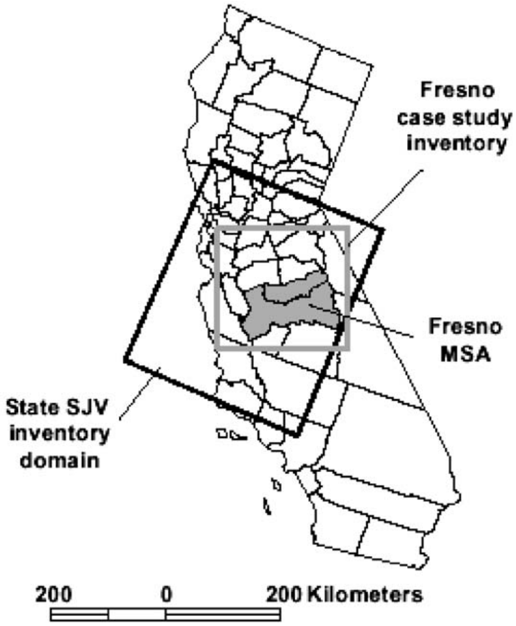

Fig. 2. Fresno area inventory domain.

2. Methods

2.1. Livestock wastes

Livestock waste emissions estimates in EPA's NET are based on emission factors recommended in a 1994 study by Battye et al. (1994), which were derived from European measurements. The North Carolina and SJV inventories are also based largely on the Battye et al. report, although the composite emission factors were recalculated using state-specific animal size distributions.

The current inventory was based on a number of studies published since 1994, which are summarized in Table 1, along with the emission factors used in the current study. Each emission factor includes total emissions resulting from animal housing, grazing, manure storage, and land spreading. The large variation in estimates illustrates the difficulty in developing precise estimates. Most of the experimental emission factors are obtained from Europe, where animal practices may vary significantly from the United States. The use of European emission factors has drawn considerable criticism from the agricultural community. In order to ground proof the European emission factors, the agricultural design references that provide estimates of the average nitrogen content of wastes produced by domestic cattle was reviewed. The Midwest Plan Service waste management handbook provides estimates of the amount of nitrogen, on average, in wastes produced by domestic cattle (Midwest Plan Service, 1993). In addition, the handbook estimates ammonia losses to the atmosphere from various waste storage and management systems. Table 1 compares emission factors developed from the Midwest Plan Service handbook with the selected emission factors. As the table shows, the emission factors are within the range indicated by the design handbook.

County level estimates of animal population were obtained from the Census of Agriculture (USDA, 1999). The Census of Agriculture provides population estimates for beef and dairy cattle; therefore, a separate emission factor was determined for each group. The 1994 EPA ammonia report recommended an emission factor of about 15 kg-NH_3/animal-yr for beef cattle or "young cattle for fattening." In the current inventory, the average of the three most recent published factors from Table 1, Buowman et al. (1997), Misselbrook et al. (2000) and Van Der Hoek (1998) were used. The resulting average emission factor is 10.2 kg-NH_3/animal-yr. A similar approach is used for dairy cattle, taking the average of emission factors given by the European Environment Agency (EMEP) (McInnes, 1996), Misselbrook et al. (2000) and Van Der Hoek (1998) to obtain a factor of 28 kg-NH_3/animal-yr.

Hogs and pigs are not divided into weight or class categories in the Census of Agriculture; however, Van Der Hoek (1998) suggests that three classes can be determined based on the total population of hogs. One can assume that approximately 50% are fattening hogs, 10% are sows, and the remaining 40% are young sows and piglets. Two separate emission factors, 6.4 and 16.4 kg-NH_3/animal-yr, are derived for fattening hogs and sows respectively. This factor for fattening hogs is in agreement with recent studies at a commercial hog farm by McCulloch et al. (1998). His study estimated total

Table 1
Summary of recent published emission factors for livestock (kg-NH_3/animal-yr).

Source	Recent estimates and measurements						Estimated from waste design handbook	Selected for the current case studies
	ECETOC (1994)	EMEP (McInnes, 1996)	Buowman et al. (1997)	UNECE (Van Der Hoek, 1998)	Misselbrook et al. (2000)	Other measurements		
Dairy cow	40	29	25	29	27	23[a]	20–70	28
Beef cow	28	15	9.5	14	6.8		9–18	10.2
Pigs	4.3	—	4.9	—	—			
Sow	—	16.6	—	16	5.2	5.9–12[b]	14–17	16.4
Finishing pig	—	6.5	—	6.4	4.8		5–10	6.4
Poultry	0.19	—	0.24	0.37	—			
Laying hen	—	0.38	—	—	0.45		0.2–0.4	0.37
Broiler	—	0.27	—	0.28	0.23		0.1–0.2	0.28
Sheep	1.8	1.5	0.77	1.3	0.73			1.34
Horses	11.9	—	9.2	8.0	—			8.0

(—) indicates that no estimates or measurements were available.
[a] Based on results of Schmidt and Winegar (1996) as modified by Winegar and published in the SCAQMD emissions inventory (Botsford et al., 1997).
[b] McCulloch et al. (1998), for finishing pigs.

NH_3 emissions from hog facilities to be in the range of 5.9–11.6 kg-NH_3/animal. Aneja et al. (2000) measured emissions from a hog waste lagoon at about 2.7 kg/animal-yr. They estimated that these emissions account for about one-third of total emissions from the hog feeding facility, resulting in overall facility emissions of about 8.0 kg-NH_3/animal) (Aneja et al., 2000).

Emission factors for sheep, broilers, and laying hens were updated based on new experimental data, given by Van Der Hoek (1998). For the remaining animal groups (pullets 13–20 weeks, pullets <30 weeks, and turkeys) and fertilizer application, the emission factors recommended in the 1994 EPA ammonia document are used.

2.2. Soils and vegetation

Measurements of ammonia emissions from forests and other uncultivated lands range over several orders of magnitude (Lenhard and Gravenhorst, 1980; Schlesinger and Hartley, 1992; Van Der Hoek, 1998). In fact, in the short term, fluxes ranged from −410 kg/ha-yr (net deposition) to +220 kg-NH_3/ha-yr (net emission) (Duyzer et al., 1994; Wyers and Erisman, 1998). Table 2 presents the emission factors selected for this case study emissions inventories. For forests, an emission factor of 1.2 kg-NH_3/ha-yr was selected. This value is at the low end of the range given by Schlesinger and Hartley (1992), and at the high end of the range cited in the recent review by Buowman et al. (1997) (Kinnee et al., 1997). The recent micrometeorological measurements by Wyers and Erisman (1998), which provide a continuous record of ammonia fluxes in a Douglas Fir forest in the Netherlands for a period of more than 2 yr were also considered (Hegg et al., 1990). They calculated net emissions at 0.14 kg/ha-yr for 1993, and 0.05 kg/ha-yr for 1994. However, the ambient ammonia concentration during these measurements was about 5 μg/m^3. At this level, ammonia concentrations may have exceeded the compensation point for a significant portion of the year. No long-term measurements of ambient concentrations are available for the US, however, REMSAD modeling predicts ammonia concentrations of 3 μg/m^3, or below, for the case study areas.

Table 2
Emission factors selected for non-agricultural soils and vegetation

Land cover	Selected emission factor (kg-NH_3/ ha-yr)	Basis
Forests	1.2	Schlesinger and Hartley (1992), Buowman et al. (1997)
Grassland (non-agricultural)	0.3	Buowman et al. (1997) and Schlesinger and Hartley (1992)
Shrub land	0.4	Buowman et al. (1997)
Barren lands	0.1	Buowman et al. (1997)
Built-up land	0.1	Calculated by applying the grassland emission factor to 33% of the land surface, as in the SJV inventory

For non-agricultural grasslands an emission factor of 0.3 kg/ha-yr was selected. This is equal to the factor given by Buowman et al. (1997), and is near the geometric mean of the range of factors recommended by Schlesinger and Hartley (1992). However, it is considerably lower than the factor used in the California SJV inventory, 5.5 kg/ha-yr. For shrub land and barren lands, Buowman's factors of 0.4 and 0.1 kg/ha-yr, respectively, were used. Agricultural land or pasture land in these emission calculations are not included, because emissions from these land uses are already covered in the livestock and fertilizer categories. Soil and vegetation emissions were calculated by applying the emission factors in Table 2 to the EPA's BELD database. The BELD database was developed by EPA in 1997 for use with its Biogenic Emissions Inventory System (BEIS) model, and includes various agricultural crops and forest species, at a 1-km resolution (Kinnee et al., 1997).

The emissions estimates for soils and vegetation are subject to a great deal of uncertainty. The difference between the current best estimate of emissions and the upper bound estimate is about an order of magnitude. An upper bound emission factor of 10 kg/ha-yr for forests was estimated, based on the upper bound given by Schlesinger and Hartley (1992). The upper bound for non-agricultural grasslands is estimated at 2 kg/ha-yr, based on the range given by Buowman et al. for non-agricultural grass lands (Van der Hoek, 1998). The lower bound estimate for soils and vegetation is zero, reflecting a condition where deposition to these systems outweighs the emission component on an annual basis.

2.3. Motor vehicles

Recent US tunnel tests and remote sensing tests have given average emission rates of 72 mg/km (Fraser and Cass, 1998), 49 mg/km (Kean et al., 2000), and 138 mg/km (Baum et al., 2000). The tunnel tests by Fraser and Cass, and Kean et al., and the remote sensing tests by Baum et al. roughly bracket the emission factor used in the NET inventory for catalyst vehicles, 85.4 mg/km. Therefore, the NET emission factors for catalyst vehicles, as well as other vehicle types were used. The emission factors were applied to county level estimates of vehicle miles traveled (VMT) from the NET inventory.

2.4. Fertilizer application

Fertilizer emission factors from Battye et al. (1994) were used in this study. These factors were used with county-level estimates of fertilizer consumption by type and grade, obtained from the Association of American Plant Food Control Officials (AAPFCO, 1999).

2.5. Industrial and combustion sources

The inventories for industrial and combustion sources in the Fresno area (the SJV) and in North Carolina are based primarily on emissions reported in the California Air Toxics Emission Data Systen (ATEDS) database and the North Carolina air toxics database in 1998. Emissions estimates for the South Carolina portion of the Charlotte domain are based on EPA's TRIS database. Spills and leaks of ammonia reported to the Coast Guard's National Response Center were also included, but these were very small in the two case study areas.

2.6. Sewage treatment

In the previous ammonia inventory for the SJV, an average ammonia emission factor was computed for sewage treatment plants based on emission reports in ATEDS (Coe et al., 1998). This calculation was repeated using an updated version of ATEDS. An emission factor was computed for each facility reporting to ATEDS, by dividing the overall ammonia emission rate by the effluent flow rate reported in the EPA Office of Water's Permit Compliance System (PCS). This calculation yielded an average emission factor of about 0.15 g-NH_3/m^3-water.

2.7. Other

Emission factors for human breath, human sweat, cigarette smoking, infant diapers, and pets were taken from Sutton et al. (2000).

Hegg et al. (1990) estimated ammonia emissions from biomass burning at about 1.81 ± 0.87 g-NH_3/kg-carbon burned, based on an analysis of gases collected from five forest fires located throughout North America (Hegg et al., 1990). This factor has been used in global emissions inventories by Schlesinger and Hartley (1992) and by Buowman et al. (1997) (Kinnee et al., 1997; Van der Hoek, 1998). This factor, which translates to 0.8 ± 0.4 g-NH_3/kg-biomass was used, to estimate emissions from forest fires and agricultural burning in the two case study areas. For forest fires, the total area burned was estimated from computerized fire incident data bases developed by the US Forest Service, the Federal Bureau of Land Management (BLM), the National Interagency Fire Center (NIFC-Boise, Idaho), and the state of North Carolina (California's fire incident database is not yet computerized). The amount of biomass burned per unit area were estimated at 40 Mg/hectare (Mg/ha) for California, and 20 Mg/ha for North Carolina, based on guidance given in EPA's AP-42 Compilation of Emission Factors (USEPA, 2000b).

3. Results and discussion

Fig. 3 illustrates the distribution of emissions among major source categories in the case study inventories. Agricultural sources accounted for the bulk of ammonia emissions in both case studies. Livestock waste contributed about 80% in North Carolina and 64% in the SJV, while fertilizer application contributed about 6–7% in both domains. Forests and non-agricultural vegetation contributed 5% in North Carolina and 12% in the SJV. Motor vehicles accounted for about 6% of ammonia emissions in North Carolina and 14% in the SJV.

3.1. Comparisons with previous estimates

Table 3 gives a summary of the revised inventories, and compares the new emissions estimates with previous inventories for the case study areas. As Table 3 shows, total ammonia emissions in the new case study inventory for North Carolina are somewhat higher than either the previous state inventory or the EPA NET inventory. Total emissions in the new inventory for the SJV region are substantially lower than in the previous state inventory estimate, but somewhat higher than in the EPA NET inventory.

3.1.1. Agricultural emissions

Emissions from livestock continue to make up the largest share of overall emissions for both the North Carolina and SJV case studies. Estimated emissions from livestock are similar to previous estimates for North Carolina, but lower than in estimates for the SJV region. Fertilizer emissions in both case study inventories are similar to previous estimates.

3.1.2. Soils and vegetation

The soils and vegetation category is the source of the largest variations among current ammonia emissions inventories. The largest change in the new case study inventories also derives from the soil and vegetation source category. Emissions in the new case study inventory for the SJV region are an order of magnitude lower than the corresponding estimate in the state of California's SJV inventory (see Table 3) (Coe et al., 1998). These differences stem from the inherent variability of ammonia emissions from these soil and vegetation. In fact, emissions from soils and vegetation

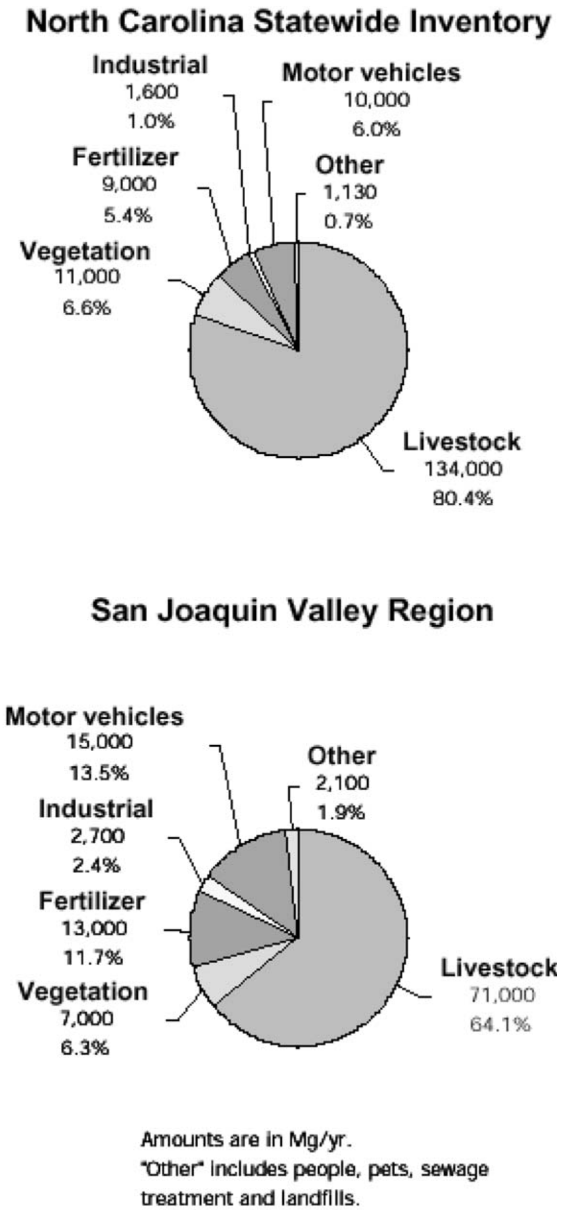

Fig. 3. Distribution of ammonia emissions among major source categories in the case study inventories (where $Mg = 10^6$).

were not included in either the EPA NET ammonia inventory or North Carolina's statewide inventory, since they can be either a source or a sink for ammonia depending on short-term conditions (NCDEHNR, 1997; USEPA, 2000b).

The difference between soil and vegetation emissions in this case study inventory and the earlier CARB SJV inventory stems primarily from differences in emission factors and in assumptions for agricultural land. An area-based emission estimate for agricultural land was not included because it was believed that such emissions would already be reflected in the estimates for livestock and fertilizer. As a result, forests were the largest source of emissions in the estimates for the soil and vegetation category.

An emission factor of 1.2 kg-NH_3/ha-yr was used for both coniferous and deciduous forests. This value is at the high end of a range cited in a recent review by Buowman et al. (1997), and higher than recent measurements in the Netherlands by Wyers and Erisman (1998) but at the low end of a range given by Schlesinger and Hartley (1992), and somewhat lower than the factor used in the previous SJV inventory (Coe et al., 1998). A wide range of emission rates measured for forests suggests that ammonia emissions will vary not only with the season, but also across any large spatial region. Ammonia emissions may be lower (or non-existent) for forests that are located in close proximity to other sources such as livestock and highways, because of a tendency to absorb ammonia from the air at high concentrations. However, short-term model predictions (or measurements) are not currently available to reliably estimate these spatial variations.

3.1.3. Motor vehicles

Table 3 shows that the current emissions estimates for motor vehicles are much higher than the previous SJV inventory (a factor of 7), and also somewhat higher than in the EPA NET inventory (Coe et al., 1998; USEPA, 2000b). The previous North Carolina statewide inventory did not include ammonia from motor vehicles (NCDEHNR, 1997).

The bulk of motor vehicle ammonia emissions emanates from vehicles equipped with three-way catalytic converters. The previous SJV inventory used an ammonia factor of 16.1 mg/km for these vehicles, based on emission tests under controlled conditions (Coe et al., 1998). The EPA NET inventory used a considerably higher emission factor, 86 mg/km (USEPA, 1998). Researchers have recently measured ammonia emissions under actual driving conditions. Average emission factors in these studies were 49 mg/km (Kean et al., 2000), 72 mg/km (Fraser and Cass, 1998), and 138 mg/km (Baum et al., 2000). On average, these factors are in rough agreement with the EPA NET emission factor for light duty vehicles equipped with three-way catalysts (86 mg/km). Therefore, that factor in the case study inventories was adopted. The increases in emissions between the EPA NET inventory and the case study inventories result from a change in base year and some differences in emission factors for heavier vehicles.

3.1.4. Other sources

The case study inventories include new emissions estimates for humans in the SJV region, and pets and

Table 3
Comparison of new case study inventories with previous inventories

Category	North Carolina statewide domain			California San Joaquin Valley domain		
	New inventory	NC agency estimates (1995)	EPA NET inventory (1998)[a]	New inventory	CARB SARMAP inventory	EPA NET inventory (1998)[a]
Emissions (1000 Mg/year)						
Livestock waste	134	138	127	71	99	89
Soils and vegetation	11	na	na	6.9	80[b]	na
Fertilizer application	9.5	8.8	9.0	13	12	13
Industrial sources	1.6	1.7	0.3	12	6.0	5.6
Motor vehicles	10	0.0	6.8	15	2.0	10
Sewage treatment plants	0.17	4.7	1.5	1.4	0.02	<0.01
Landfills	na	na	na	0.37	<0.01	na
Wild animal wastes	na	na	na	na	na	na
Human beings	0.20	1.7	na	0.35	na	na
Pets	0.76	na	0.0	1.4	na	0.0
Forest fires	0.50	na	na	5.5	na	na
Total	167	155	145	127	200	119
Distribution of emissions (%)						
Livestock waste	80	89	88	56	50	75
Soils and vegetation	6.3	na	na	5.4	40 [b]	na
Fertilizer application	5.7	5.7	6.2	11	6.2	11
Industrial sources	1.0	1.1	0.2	9.6	3.0	4.7
Motor vehicles	6.1	na	4.7	12	1.0	8.5
Sewage treatment plants	0.1	3.0	1.0	1.1	0.01	<0.01
Landfills	na	na	na	na	<0.01	na
Wild animal wastes	na	na	na	na	na	na
Human beings	0.1	1.1	na	0.3	na	na
Pets	0.5	na	0.3	1.1	na	0.7
Forest fires	0.3	na	na	4.3	na	na

na—not broken out separately, or not estimated.
[a] Data extracted from the National Emissions Trends (NET) inventory for the North Carolina and the San Joaquin Valley.
[b] The soils and vegetation category for the San Joaquin Valley and South Coast inventories includes emissions from agricultural land.

forest fires in both regions. Separate estimates have not been developed for wild animal waste, because it was felt that such estimates would double-count emissions in the soils and vegetation category.

3.2. Spatial and temporal distribution of ammonia emissions

The distribution of emissions among source categories in urban areas is less heavily weighted toward agricultural sources and more heavily weighted toward highway vehicles, as shown in Fig. 4. Figs. 5 and 6 show the seasonal distributions of the ammonia emissions estimates for the state of North Carolina as a whole and in the region around Charlotte (Mecklenburg county), respectively. Figs. 7 and 8 show seasonal distributions of our emissions estimates for the entire SJV domain (defined based on the CARB SJV inventory), and the region around the city of Fresno (about 400 km^2). As the figures show, emission estimates for agricultural sources (livestock and fertilizer application) decline to approximately 14% in the winter for both the Charlotte and Fresno urban areas. Emissions estimates for soils and vegetation also decline to approximately 0 during the winter for both the Fresno and Charlotte area. As a result, motor vehicles account for a larger fraction (approximately 73% and 70% for Charlotte and Fresno respectively) of winter ammonia emissions, particularly in the Charlotte urban area.

3.3. Uncertainties of emissions inventory estimates

Figs. 9 and 10 illustrate best, upper and lower estimates for the North Carolina and SJV case study emissions inventories, respectively. Upper and lower estimates are based on the highest and lowest emission factors available in the recently published literature (Table 1). In terms of the potential change in mass of

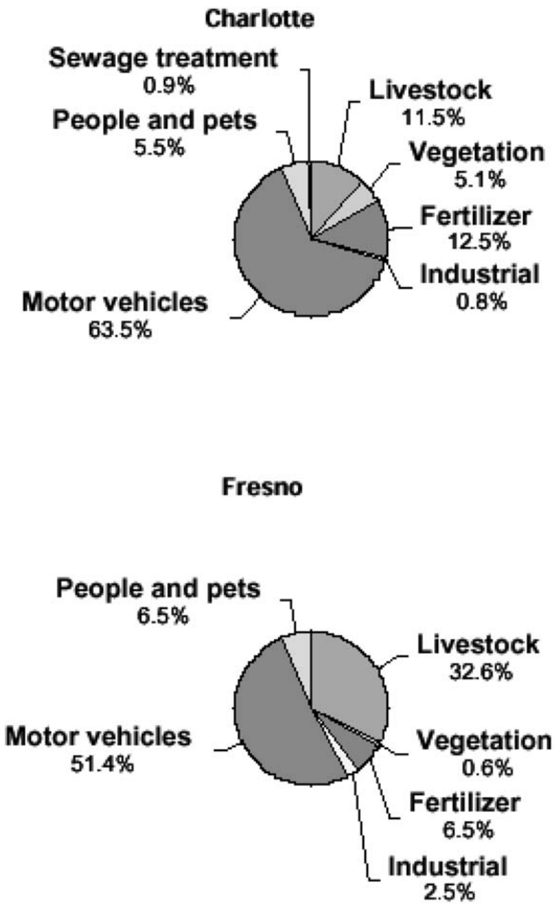

Fig. 4. Source category distribution of ammonia emissions in the immediate vicinities of Charlotte and Fresno. The areas represented in this figure are for Charlotte, Mecklenburg County; and grids covering the city of Fresno, approximately 400 km^2.

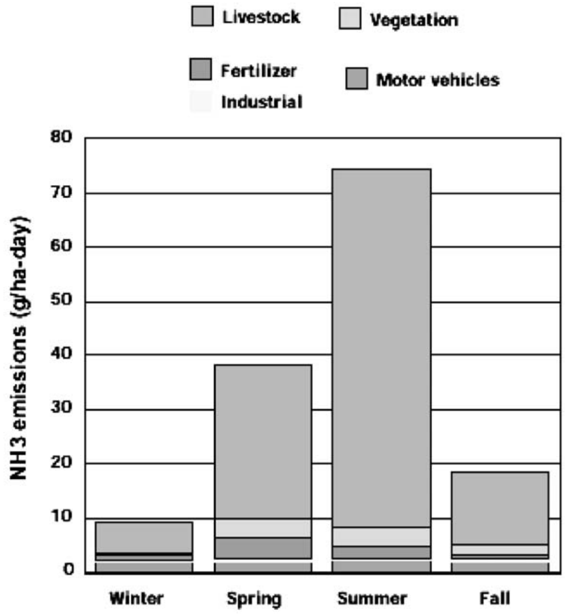

Fig. 5. Seasonal distribution of new emissions estimates for North Carolina.

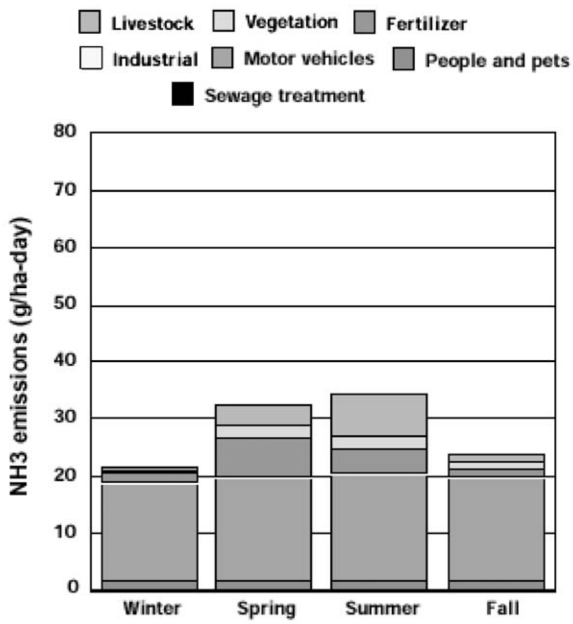

Fig. 6. Seasonal distributions of new emissions estimates for the immediate vicinity of Charlotte (Mecklenburg County).

ammonia emissions over the entire regional domains, the uncertainty for soils and vegetation, and livestock waste represents approximately 225,000 Mg NH_3/yr. All of these uncertainties stem from variations in measured emission factors. The derivations of uncertainty estimates for individual source categories are discussed in the following sections.

All of the sources of ammonia emissions are subject to variability and uncertainty. From a standpoint of aerosol modeling, the soils and vegetation category represents one of the most important sources of uncertainty. Plants will either absorb or give off ammonia, depending on the concentration of an ammonium ion in the plant and the concentration of ammonia gas in the surrounding air (Warneck, 1988). Therefore, the emissions estimates used in atmospheric models for vegetation should probably be linked to predicted concentrations of free ammonia, which would vary through the year and across the modeling domain.

The lack of monitoring data on ammonia gas is another important data gap. Without data on ammonia

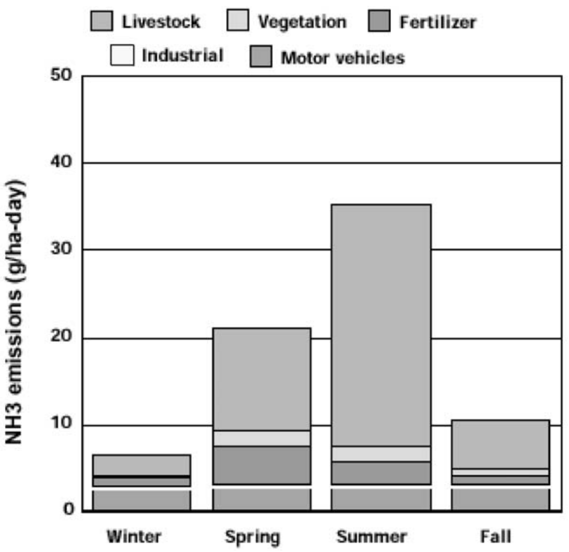

Fig. 7. Seasonal distribution of new emissions estimates for the San Joaquin valley region (as defined in the CARB SJV inventory).

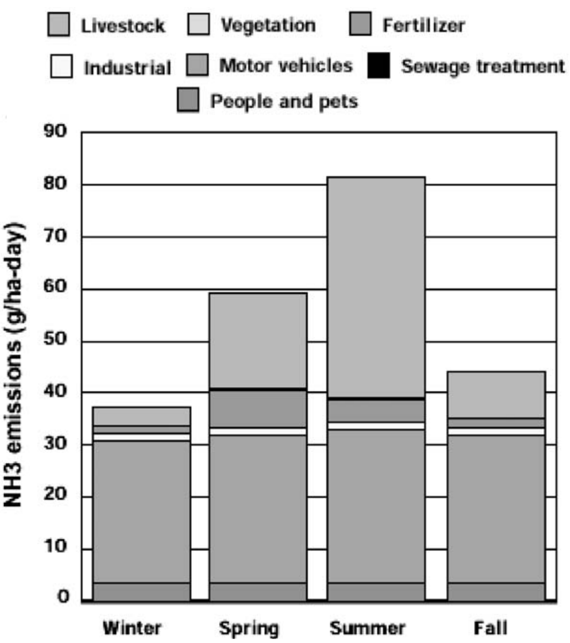

Fig. 8. Seasonal distribution of new emissions estimates for the immediate vicinity of Fresno (grids covering the city of Fresno, approximately 400 km²).

gas, there is little that can be done to evaluate the validity of the ammonia inventory. This lack of data also complicates any evaluations of modeled equilibria among NH_3, $NH4^+$, NO_3^- and $SO4^{2-}$.

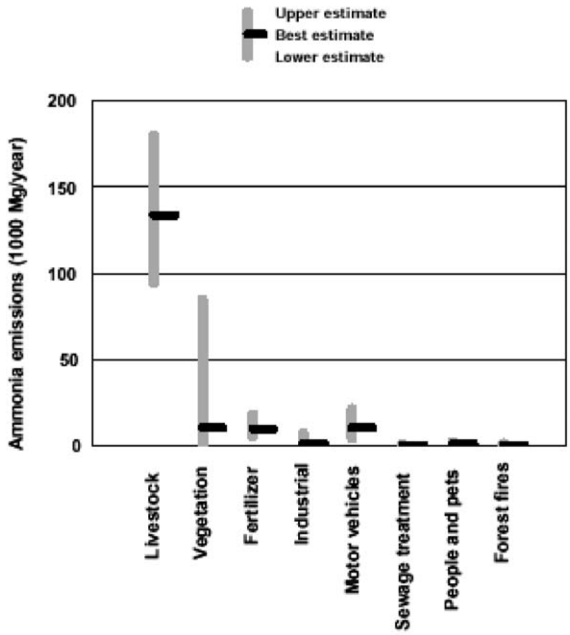

Fig. 9. Uncertainties estimates for the Charlotte/North Carolina case study inventory. Upper and lower estimates are based on the highest and lowest emission factors available in the recently published literature (Table 1).

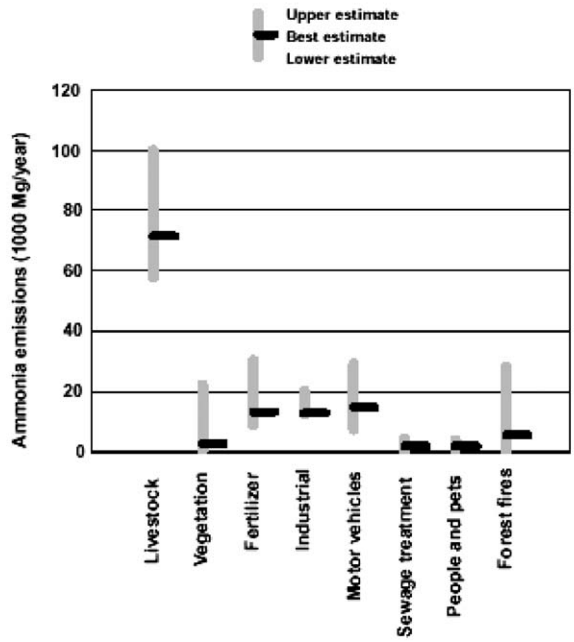

Fig. 10. Uncertainties estimates for the Fresno/SJV case study inventory. Upper and lower estimates are based on the highest and lowest emission factors available in the recently published literature (Table 1).

In terms of the mass of emissions, uncertainties for the livestock waste category are very large. However, livestock emissions have been subject to a good deal of recent attention, and the body of emissions data for that category will increase substantially in the near future as a result of ongoing measurement programs by USDA, EPA, and others (Aneja et al., 2001). In addition, livestock emissions are highest in rural areas and in the summer. In both of these situations, changes in the concentration of ammonia gas may not have a strong impact on the formation of aerosol ammonium particulates (Ansari and Pandis, 1998). Uncertainties in ammonia emissions would have the strongest impact in winter, when the formation of NH_4NO_3 particulate is favored by colder temperatures, and in urban areas, where ammonia may be the limiting component in the formation of particulate NH_4NO_3.

In the winter and in urban areas, the largest uncertainties are in ammonia emissions from motor vehicles. Recent measurements of average ammonia emissions under actual driving conditions vary by a factor of 2. However, the largest unknown is the ammonia emission factor for future catalyst systems. The ammonia emission rate is not a design criterion for automobile catalyst systems. Average emissions of ammonia have increased in the past decade with the adoption of three-way catalyst technology, and recent tests show that ammonia emissions can be much higher than these average values under some conditions. Thus, it is possible that ammonia emissions would increase even further for new catalysts.

Additional work is also needed in the temporal allocation of emissions from livestock waste and commercial fertilizers. Although additional measurements would be helpful, a better characterization of the timing of waste and fertilizer application would also reduce the uncertainty of seasonal emissions estimates.

Acknowledgements

This research was supported by the Coordinating Research Council (CRC), 3650 Mansell Road, Suite 140, Alpharetta, Georgia 30022, under CRC Contract No. A-32. Members of the CRC, in particular CRC Atmospheric Impacts Committee, provided important guidance during the preparation of this report. The authors also wish to acknowledge contributions from Steve Fudge, Rebecca Battye, and Dan Bowman of EC/R Incorporated, and Dena Nelson of North Carolina State University.

Disclaimer

Although the report has been reviewed by the CRC, the opinions, findings, and conclusions expressed are those of the authors, and not necessarily those of the CRC. Mention of trade names or commercial products does not constitute endorsement or recommendation for their use.

References

Aneja, V.P., Murray Jr., G., Southerland, J., 1998. Atmospheric nitrogen compounds: emissions, transport, transformation, deposition and assessment. Environmental Manager—Air and Waste Management Association, April, pp. 22–25.

Aneja, V.P., Chauhan, J.P., Walker, J.T., 2000. Characterization of atmospheric ammonia emissions from swine waste storage and treatment lagoons. Journal of Geophysical Research—Atmospheres 105, 11535–11545.

Aneja, V.P., Roelle, P.A., Murray, G.C., Southerland, J., Erisman, J.W., Fowler, D., Asman, W., Patni, N., 2001. Atmospheric nitrogen compounds II: emissions, transport, transformation, deposition and assessment. Atmospheric Environment 35, 1903–1911.

Aneja, V.P., Nelson, D.R., Rolle, P.A., Walker, J.T., Battye, W., 2003. Agricultural ammonia emmissions and ammonium concentrations associated with aerosols and precipitation in the southeast United States. Journal of Geophysical Research 108, 4152–4162.

Ansari, A.S., Pandis, S.N., 1998. Response of inorganic PM to precursor concentrations. Environmental Science and Technology 32, 2706–2714.

Association of American Plant Food Control Officials, Inc.—AAPFCO, 1999. Commercial Fertilizers, 1998. Lexington, KY.

Battye, R., Battye, W., Overcash, C., Fudge, S., 1994. Development and selection of ammonia factors. EPA/600/R-94/190, Final Report Prepared for US Environmental Protection Agency, Office of Research and Development, EPA Contract No. 68-D3-0034, Work Assignment 0–3.

Baum, M.M., et al., 2000. Remote sensing of criteria and noncriteria pollutants emitted in the exhaust of on-road vehicles. 220th American Chemical Society National Meeting. August 20–24, Washington, DC.

Botsford, C., Chitjian, M., Koizumi, J., Yijin Wang, Gardner, L., Winegar, E., 1997. 1997 Gridded Ammonia Emission Inventory Update for the South Coast Air Basin. 52.16156.5204. Prepared by Systems Applications International for the South Coast Air Quality Management District.

Buowman, A.F., Lee, D.S., Asman, W.A.H., Dentener, F.J., Van Der Hoek, K.W., Olivier, J.G.F., 1997. A global high-resolution emission inventory for ammonia. Global Biogeochemical Cycles. 11 (4), 561–587.

Coe, D., Chinkin, L., Loomis, C., Wilkinson, J., Zwicker, J., Fitz, D., Pankratz, D., Ringler, E., 1998. Technical Support Study 15: Evaluation and Improvement of Methods for Determining Ammonia Emissions in the San Joaquin Valley. Prepared by Sonoma Technology, Inc., Santa Rosa, California for the California Air Resources Board, Sacramento, CA.

Duyzer, J.H., Verhagen, H.L.M., Westrate, J.H., Bosveld, F., Vermetten, A.W.M., 1994. The dry deposition of ammonia onto a douglas fir forest in the Netherlands. Atmospheric Environment 28, 1241–1253.

European Centre for Ecotoxicology and Toxicology of Chemicals—ECETOC, 1994. Ammonia emissions to air in Western Europe. Technical Report No. 62, Brussels, Belgium.

Fraser, M.P., Cass, G.R., 1998. Detection of excess ammonia emissions from in-use vehicles and implications for fine particle control. Environmental Science and Technology 32 (8), 1053–1057.

Hegg, D.A., Radke, L.F., Hobbs, P.V., Rasmussen, R.A., Riggan, P.J., 1990. Emissions of some trace gases from biomass fires. Journal of Geophysical Research 95, 5669–5675.

Kean, A.J., Harley, R.A., Littlejohn, D., Kendall, G.R., 2000. On-road measurement of ammonia and other motor vehicle exhaust emissions. Environmental Science and Technology 34 (17), 3535–3539.

Kinnee, E., Geron, C., Pierce, T., 1997. US land use inventory for estimating biogenic ozone precursor emissions. Ecological Applications 7, 46–58.

Lenhard, U., Gravenhorst, G., 1980. Evaluation of ammonia fluxes into the free atmosphere over western Germany. Tellus 32, 48–55.

McCulloch, R.B., Few, G.S., Murray, G.C., Aneja, V.P., 1998. Analysis of ammonia, ammonium aerosols and acid gases in the atmosphere at a commercial hog farm in Eastern North Carolina, USA. Environmental Pollution 102, 263–268.

McInnes, G. (Ed.), 1996. Joint EMEP/CORINAIR Atmospheric Emission Inventory Guidebook. Vol. 1. European Environment Agency, Copenhagen, Denmark.

Midwest Plan Service, 1993. Livestock Waste Facilities Handbook. Iowa State University, Ames, IA.

Misselbrook, T.H., Van Der Weerden, T.J., Pain, B.F., Jarvis, S.C., Chambers, B.J., Smith, K.A., Phillips, V.R., Demmers, T.G.M., 2000. Ammonia emission factors for UK agriculture. Atmospheric Environment 34, 871–880.

North Carolina Department of Environment, Health and Natural Resources (NCDEHNR), 1997. Assessment Plan for Atmospheric Nitrogen Compounds: Emissions, Transport, Transformation, and Deposition. March 10.

Schlesinger, W.H., Hartley, A.E., 1992. A global budget for atmospheric NH_3. Biogeochemistry 15, 191–211.

Schmidt, C.E., Winegar, E., 1996. Results of the measurements of PM_{10} precursor compounds from dairy industry livestock waste. Technical Report Prepared for the South Coast Air Quality Management District.

Sutton, M.A., Dragosits, U., Tang, Y.S., Fowler, D., 2000. Ammonia emissions from non-agricultural sources in the U.K. Atmospheric Environment 34, 855–869.

US Environmental Protection Agency (USEPA), 1998. National Air Pollutant Emission Trends, Procedures Document, 1900–1996, EPA-454/R-98-008. Office of Air Quality Planning and Standards, Research Triangle Park, NC.

US Environmental Protection Agency (USEPA), 2000a. Compilation of Air Pollutant Emission Factors, AP-42, Vol. I—Stationary Sources. Research Triangle Park, NC.

US Environmental Protection Agency (USEPA), 2000b. National Air Pollutant Emission Trends, 1900–1988, EPA-454/R-00-002. Research Triangle Park, NC.

United States Department of Agriculture (USDA), 1999. 1997 Census of Agriculture. Vol. 1, Part 1 (Chapter 2).

Van Der Hoek, K.W., 1998. Estimating ammonia emission factors in Europe: summary of the work of the UNECE ammonia expert panel. Atmospheric Environment 32, 315–316.

Warneck, P., 1988. Chemistry of the Natural Atmosphere. Academic Press, San Diego, CA.

Wyers, G.P., Erisman, J.W., 1998. Ammonia exchange over coniferous forest. Atmospheric Environment 32, 441–451 (Ammonia Special Issue).