

Emissions Inventory Guidance for Anthropogenic Non-Agricultural Ammonia Sources

Stephen M. Roe, Holly C. Lindquist, Kirstin B. Thesing, Melissa D. Spivey, Randy P. Strait
E.H. Pechan & Associates, Inc., P.O. Box 1345, El Dorado, CA 95623

sroe@pechan.com

holly.lindquist@pechan.com

kthesing@pechan.com

melissa.spivey@pechan.com

rstrait@pechan.com

Roy Huntley

U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Emission Modeling and Analysis Division, Emission Factor and Inventory Group,
Research Triangle Park, NC 27711

Huntley.Roy@epamail.epa.gov

Abstract

New Emissions Inventory Improvement Program (EIIP) guidance on the development of emission inventories for anthropogenic sources of ammonia (NH₃) has been developed (Roe et al, 2004a). The purpose of this new emissions guidance for “anthropogenic sources” is to update the materials presented in a 1994 U.S. Environmental Protection Agency (EPA) report on estimating ammonia emissions (Battye et al, 1994). Updated guidance is available for estimating ammonia emissions from industrial sources, combustion sources, and miscellaneous sources. For the purposes of this guidance, the term “anthropogenic sources,” excludes emissions from the agricultural sector (e.g., fertilizer application, livestock operations), as well as natural sources (e.g., soils, wild animal populations). As compared to dominant NH₃ sources such as livestock operations, the anthropogenic sources covered in this guidance are estimated to contribute small amounts to national and regional annual inventories. However, at smaller spatial and temporal scales (e.g., urban), the sources covered in this guidance can make significant contributions to an ammonia emissions inventory. The scope of this work was limited to the identification and documentation of readily-available emissions data. A literature review was performed; however, no testing programs were conducted in support of this project. The primary objective of this guidance is to provide updated information to developers of regional NH₃ inventories in support of air quality modeling.

Introduction

Ammonia (NH₃) emissions are an important contributor to fine particulate matter (PM) formation. Consequently, increased attention is being paid to accurate quantification and characterization of NH₃ emissions. A 1994 United States (U.S.) Environmental Protection Agency (EPA) report entitled *Development and Selection of Ammonia Emission Factors*, hereafter referred to as the “1994 guidance,” contains the results of a literature review and compiled NH₃ emission factors (Battye et al., 1994). The purpose of this new emissions

guidance for “anthropogenic sources” is to update the materials presented in Chapters 4 (Ammonia Emissions in Industry), 5 (Ammonia Emissions from Combustion), and portions of Chapter 6 (Miscellaneous Sources) of the 1994 guidance. For the purposes of this guidance, the term “anthropogenic sources,” excludes emissions from the agricultural sector (e.g., fertilizer application, livestock operations), as well as natural sources (e.g., soils, wild animal populations).

As compared to dominant NH₃ source sectors such as livestock operations, the anthropogenic sources covered in this guidance are estimated to contribute small amounts to national and regional annual inventories. On the other hand, as compared to natural sources and the agriculture sector, the categories covered here are often located in suburban and urban areas. Based on the current state of NH₃ emission inventories, emissions in urbanized areas are often under-represented. Figures 1 through 3 provide pie charts of ammonia source sector contributions to EPA’s National Emission Inventory (NEI), the Mid-Atlantic Northeast Visibility Union (MANE-VU) Regional Planning Organization (RPO), and the New York City Metropolitan Area (NYMA; six counties surrounding New York City, including northern New Jersey; Roe et al, 2004b).

As shown in Figures I-1 through I-3, source contributions can vary substantially depending on the spatial scale involved as well as the general land use patterns in any given area. The NEI estimates in Figure 1 were taken from the 2002 area source NEI estimates and the 1999 point, onroad, and nonroad estimates. These national estimates are dominated by the agricultural sector with about 85% contributed by livestock operations and agricultural fertilizer losses. Currently, the NEI does not include emission estimates for all categories included in this guidance, including composting and landfills. Given, the uncertainty in natural soil emission estimates, these have been excluded from all three figures.

Compared to the national-scale estimates, the source sector contributions shown in Figure 2 for the MANE-VU RPO are less dominated by the agricultural sector (about 64%). Notably, the onroad sector contributes nearly 17%. As one would expect in an urban domain, agricultural sector contributions are small. Figure 3 for the NYMA shows these contributions to be less than 5%. In this example urban area, the onroad sector contributes nearly half of the inventory. Other source sectors that can take on significance in urban areas include composting, publicly-owned treatment works (POTWs)/biosolids management, industrial sources, and domestic sources. Therefore, emission estimation methods and data sources are needed to gain a better understanding of air pollutant formation in urban areas and a more comprehensive picture of all ammonia sources. The Emission Inventory Improvement Program (EIIP) has funded the development of this guidance with these needs in mind.

Industrial, Commercial, and Municipal Source Emission Inventory Guidance

Figure 4 provides the table of contents for the new EIIP guidance (Roe et al, 2004a). The first introductory chapter is followed by a chapter on industrial, commercial, and municipal source including the following source sectors:

- *Industrial Refrigeration Losses* – fugitive losses of ammonia refrigerant;

- *Sewage Treatment* – including emissions from wastewater treatment, biosolids treatment, and biosolids management (land application);
- *Composting* – both ammonia and volatile organic compounds (VOC) emissions from the composting of biosolids, mixed wastes, and green wastes;
- *Bakeries* – emissions that occur through the use of ammonium bicarbonate as a leavening agent;
- *Pulp and Paper* – emission factors for Kraft pulping processes;
- *Surface Coatings* – ammonia is added to water-based coatings in trace quantities for pH adjustment;
- *Municipal Solid Waste Landfills* – no new emissions data were identified; however an emission factor adopted from European studies was included in the guidance;
- *Portland Cement Kilns* – cement kilns can be large sources of ammonia due depending on the nitrogen content of the kiln feed materials;
- *Accidental Releases* – as reported to the National Response Center; and
- *Miscellaneous Sources* – these include human respiration and perspiration, cigarette smoking, infant diapers, household cleaners, non-agricultural fertilizer usage.

Also included in this chapter are available ammonia emission factors from EPA's AP-42 and a number of emission factors from the California Air Resources Board's Emission Inventory Data Acquisition and Retrieval (CEIDARS) database. These emission factors include a wide range of source sectors not included above.

Combustion Source Emission Inventory Guidance

Stationary Sources

Little has changed since the 1994 guidance was prepared on the availability of NH₃ emissions data from fossil fuel-fired boilers and other stationary combustion sources. Recommended emission factors are largely taken from the 1994 guidance (Battye et al, 1994). The reported emission factors for coal combustion cover a range of four orders of magnitude (0.000565 to 2.0 lbs/ton). The emission factor recommended for utility, industrial, commercial, and institutional coal combustion in the 1994 guidance is the low end of this range and is taken from the 1985 National Acid Precipitation Assessment Program.

A discussion on ammonia emissions from nitrogen oxides control technologies using ammonia injection is also provided. The technologies are selective catalytic reduction and selective non-catalytic reduction. Different methods for estimating ammonia emissions are provided depending on the data available to the user.

Mobile Sources

Emissions data for both onroad and nonroad engines are included. A discussion of several onroad source testing programs is included. Three-way catalyst-equipped gasoline vehicles contribute most of the emissions for the onroad sector. The increasing penetration of these vehicles over the last decade has made the onroad sector one of the more important source

sectors, especially in urbanized areas. The EIIP-recommended emission factors are those used in EPA's MOBILE6.2 model.

For nonroad sources, no ammonia test data were identified. Recommended emission factors are taken from test programs for onroad engines. Emission factors for both gasoline- and diesel-powered equipment are included.

Biomass Combustion

Methods are provided in the new guidance to estimate emissions from biomass combustion. Most of the ammonia emissions occur during the smoldering (flameless) stage of combustion, when oxygen is limited. Emission factors are presented for forest fuels (i.e. wood and forest debris) and grasses/sage. The emission factors are presented in terms of either the amount of mass consumed or based on the molar ratio of NH₃ to carbon monoxide.

Conclusions

As shown in the introduction, anthropogenic sources are generally small, in terms of annual emissions, yet numerous and cover a wide range of source sectors. This differs from natural and agricultural sources, which are characterized by relatively few source sectors and produce significant levels of emissions. Intuitively, it is the anthropogenic non-agricultural source sector that is the most important to characterize in situations where air quality in urbanized areas is being evaluated (e.g., fine PM formation). This concept is demonstrated in the pie charts shown in Figures 1 through 3.

For the most part, available emission factors for anthropogenic sources are based on few measurements and have a high degree of uncertainty. Research in the following areas would help in filling data gaps or in reducing the uncertainties in anthropogenic NH₃ emissions:

- *Additional characterization of emissions from biosolids processes at POTWs and biosolids management activities:* additional measurements (e.g., through remote sensing or other techniques) are needed to provide a better understanding of emissions for various biosolids dewatering processes (e.g., sludge presses, sludge drying beds), and biosolids composting and land application;
- *Studies to identify and characterize additional emission sources in municipal sewage treatment systems:* specifically, identification and measurements of potential emission points outside of the POTW (e.g., pump/lift stations, vents);
- *Further evaluation of potentially important industrial, commercial, and municipal source categories:* for example, emissions from active MSW landfills are highly uncertain and potentially important at regional scales. In addition to the ammonia content of landfill gas, surface measurements are needed for landfills using different daily cover materials (e.g. green waste, sewage sludge); also, cement kilns can be important point sources of ammonia emissions, yet the available emissions data vary over 3 orders of magnitude (0.002 to 1.29 lb/ton of clinker). Additional emission measurements are needed in

addition to measurements of process data to develop an emission factor equation that takes into account the nitrogen content of the kiln feed materials;

- *Continued study and refinement of methods for estimating emissions from industrial refrigeration:* While direct surveys of sources that use ammonia-based refrigeration systems will continue to be the most accurate method for estimating emissions, this method is difficult to perform for large States or regions. The default employment-based emission factor provided in this guidance is based on a study in the South Coast Air Quality Management District of California, and its applicability to other areas of the U.S. is uncertain. This emission factor is thought to provide emission estimates accurate within an order of magnitude of the real value;
- *Additional information on the ammonia content of water-based surface coatings (especially architectural coatings):* very little information is available for the amount of ammonia used for pH adjustment in water-based coatings. The available information suggests that ammonia emissions might not be significant from surface coatings; however more survey data are needed for verification; and
- *Additional study of biomass combustion:* currently, available information is taken from studies on wildfires and prescribed burns. Additional measurements for agricultural burns and residential wood combustion are needed.

References

Battye et al., 1994. R. Battye, W. Battye, C. Overcash, and S. Fudge, *Development and Selection of Ammonia Emission Factors*, prepared for U.S. Environmental Protection Agency, Office of Research and Development. August 1994.

Roe et al, 2004a. Roe, S.M., M.D. Spivey, H.C. Lindquist, K.B. Thesing, R.P. Strait, *Estimating Ammonia Emissions From Anthropogenic Sources, Draft Report*, prepared for the U.S. EPA, Emission Factor and Inventory Group. March 18, 2004.

Roe et al, 2004b. Roe, S.M., Y.K. Hsu, and H.C. Lindquist, *Technical Memorandum: MANE-VU 2002 Ammonia Emissions Inventory for Miscellaneous Sources*, prepared by E.H. Pechan & Associates, Inc., prepared for the Mid-Atlantic - Northeast Visibility Union. March 2004.

Acknowledgements

The authors received valuable oversight and review from an Emission Inventory Improvement Program Technical Committee consisting of:

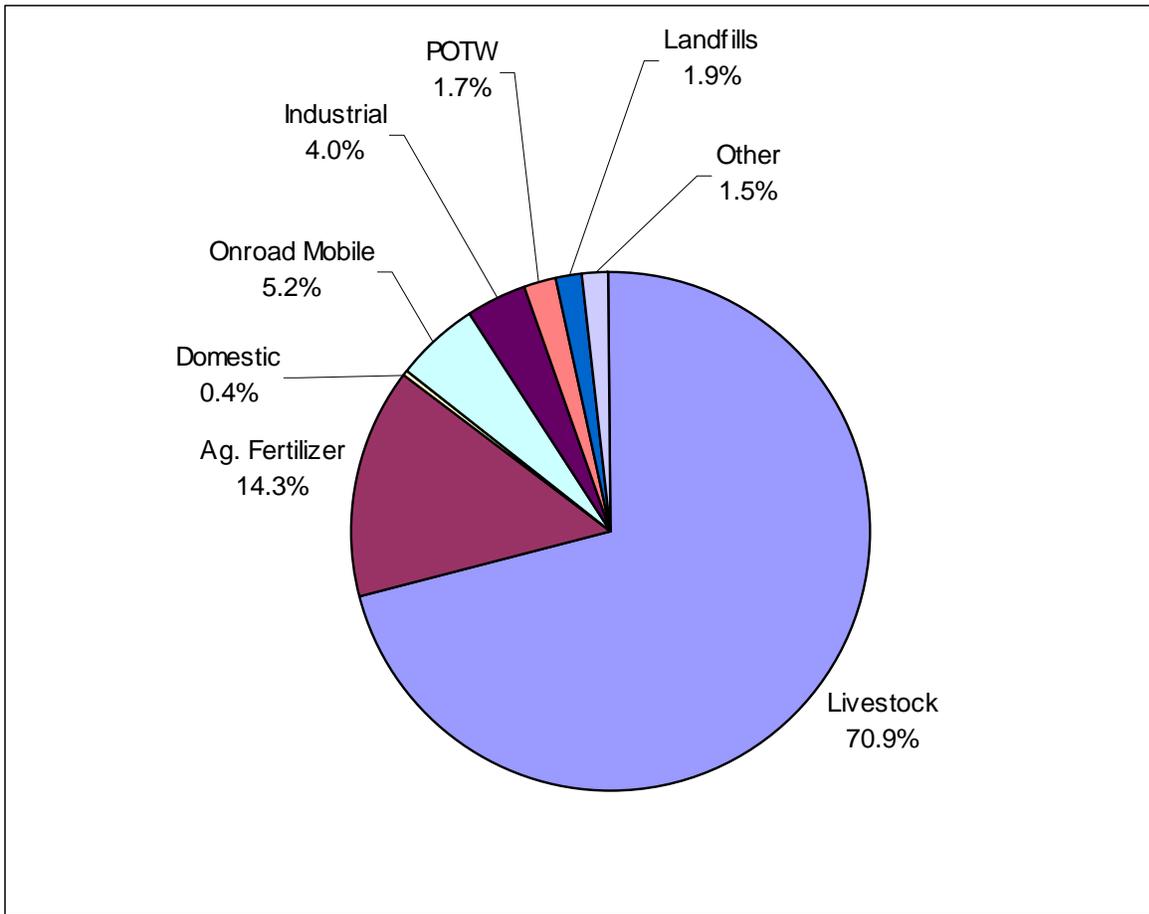
Steve Anderson - Texas Commission on Environmental Quality
Gary Beckstead - Illinois Environmental Protection Agency
Bob Betterton - South Carolina Department of Health and Environmental Control
Julia Lester - South Coast Air Quality Management District

Jim Southerland - North Carolina Department of Environment and Natural Resources
Bob Wooten - North Carolina Department of Environment and Natural Resources

The authors like to thank the following individuals or organizations for contributing information or reviewing portions of this report:

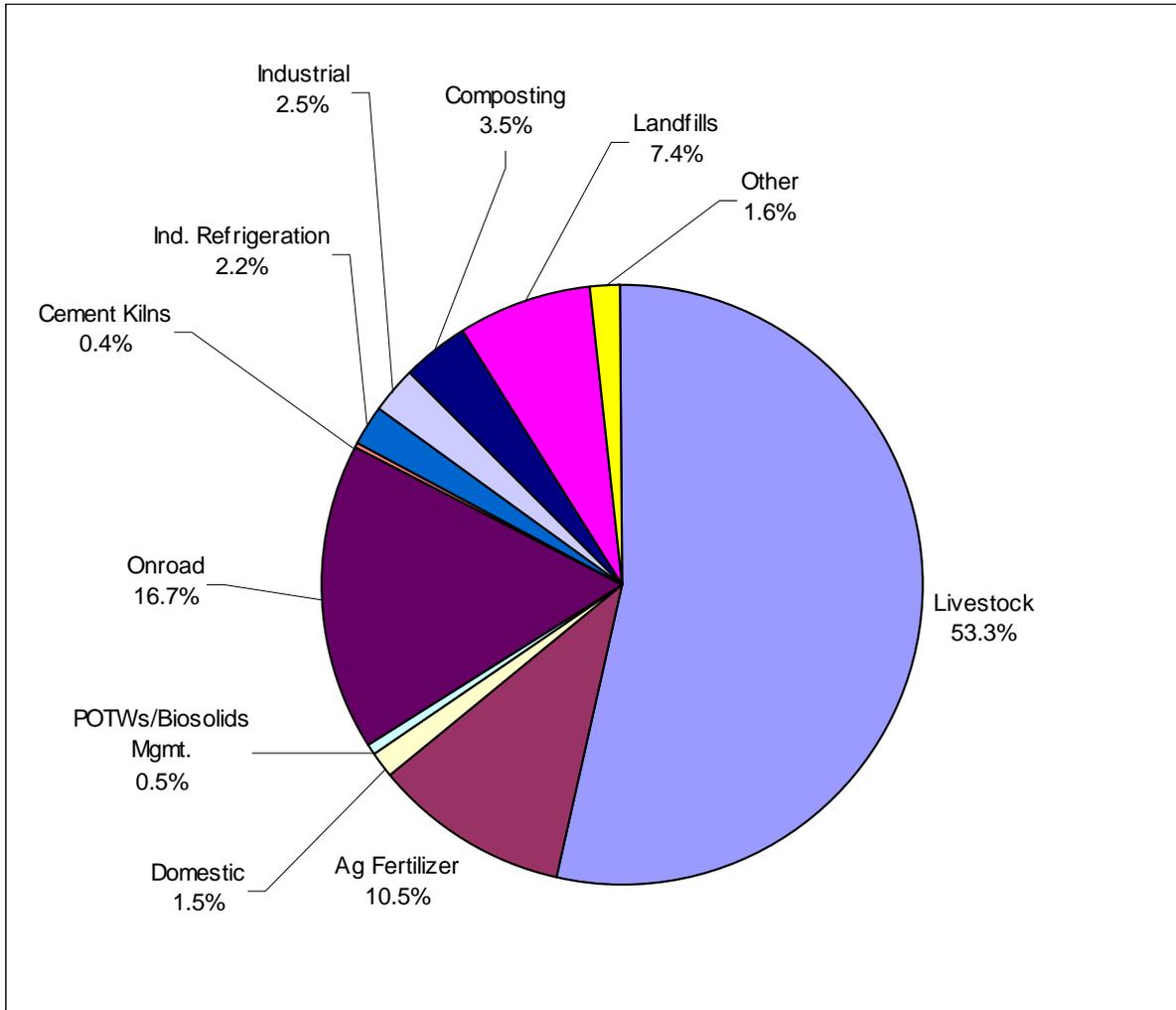
- Ms. Brenda Smyth of the California Integrated Waste Management Board - composting;
- Mr. Garth Hawkins of the Portland Cement Association - cement kilns; and
- The Western States Petroleum Association - oil and gas production and refining.

Figure 1. Source Sector-Level Ammonia Contributions to U.S. National Emissions.



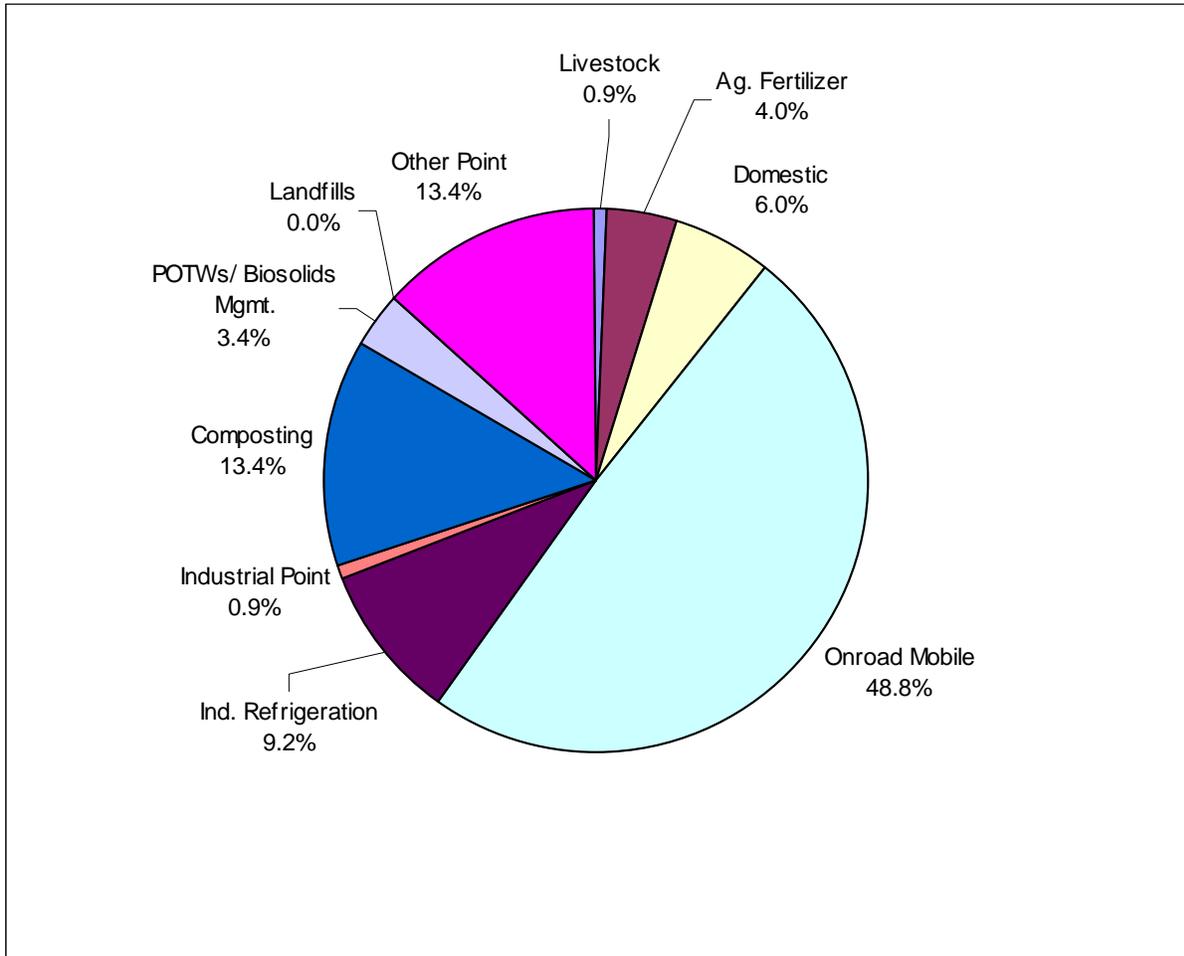
Note: Area source data are from the 2002 NEI; point, onroad, and nonroad data are from the 1999 NEI. Domestic and landfill emissions were added to the NEI emission estimates. "Domestic" includes human perspiration/respiration, infant diapers, cigarette smoke, domestic fertilizer use, and household ammonia use. "Other Point" includes non-industrial point sources. "Other" includes: nonroad mobile, commercial and residential fuel combustion, forest fires and prescribed burning, storage and transport, incineration, and industrial waste water treatment.

Figure 2. Source Sector-Level Ammonia Contributions to the MANE-VU EI.



Note: Source: Roe et al, 2004. "Domestic" includes human perspiration/respiration, infant diapers, cigarette smoke, domestic fertilizer use, and household ammonia use. "Other Point" includes non-industrial point sources. It is not clear the degree of overlap between the point source inventory and industrial refrigeration emission estimates. "Other" includes: nonroad mobile, commercial and residential fuel combustion, forest fires and prescribed burning, storage and transport, incineration, and industrial waste water treatment.

Figure 3. Source Sector-Level Ammonia Contributions to the NYMA EI.



Note: Source: Roe et al, 2004. The NYMA was defined as six New York and four New Jersey counties (Bronx, Kings, Nassau, New York, Queens, Richmond, Bergen, Essex, Hudson, and Union counties). "Domestic" includes human perspiration/respiration, infant diapers, cigarette smoke, domestic fertilizer use, and household ammonia use. "Other Point" includes non-industrial point sources. It is not clear the degree of overlap between the point source inventory and industrial refrigeration emission estimates. "Other" includes: nonroad mobile, commercial and residential fuel combustion, forest fires and prescribed burning, storage and transport, incineration, and industrial waste water treatment. Data were not available to estimate landfill emissions.

Figure 4. Table of Contents for the New EIIP Guidance.

ACKNOWLEDGMENTS

TABLES

FIGURES

ACRONYMS AND ABBREVIATIONS

CHAPTER I. INTRODUCTION

CHAPTER II. INDUSTRIAL, COMMERCIAL, AND MUNICIPAL SOURCES

- A. INDUSTRIAL REFRIGERATION LOSSES
- B. EMISSIONS FOR SPECIFIC INDUSTRIAL, COMMERCIAL, AND MUNICIPAL PROCESSES
 - 1. Sewage Treatment
 - 2. Composting
 - 3. Bakeries
 - 4. Pulp and Paper
 - 5. Surface Coatings
 - 6. Municipal Solid Waste Landfills
 - 7. Portland Cement Kilns
- C. ACCIDENTAL RELEASES
- D. MISCELLANEOUS SOURCES

CHAPTER III. COMBUSTION SOURCES

- A. EMISSIONS FROM STATIONARY COMBUSTION SOURCES
- B. AMMONIA SLIP EMISSIONS FROM NO_x-CONTROLLED COMBUSTION SOURCES
- C. MOBILE SOURCES
 - 1. Onroad Mobile Sources
 - 2. Nonroad Mobile Sources
- D. BIOMASS COMBUSTION

CHAPTER IV. RECOMMENDATIONS FOR FUTURE WORK

CHAPTER V. REFERENCES

APPENDIX A. 2002 AMMONIA ACCIDENTAL RELEASE DATA FROM THE NATIONAL RESPONSE CENTER