Assessment of Emissions Inventory Needs For Regional Haze Plans

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ABSTRACT

The Mid-Atlantic/Northeast Visibility Union (MANE-VU), and The Visibility Improvement State and Tribal Association of the Southeast (VISTAS), sponsored a project to review the existing methodologies and air emissions inventory estimates suitable for use in regional haze modeling in the Eastern United States.

The goals of the project were to:

- Explain the need for a new regional inventory to address the causes and potential control options for regional haze,
- Discuss the strengths and weaknesses of current methods used to develop inventories of regional haze precursors,
- Identify and prioritize activities that could be completed within a five year initial planning period to improve the methods and inventory, and
- Develop a list of activities, organized by year, that would serve as a guide for a work plan to be implemented by the RPOs

This paper discusses the approach used to evaluate and prioritize emissions inventory needs, discusses those specific needs that can best be completed by RPOs or individual State/Tribal/Local agencies, and presents the work plan tasks.

INTRODUCTION

The U.S. Environmental Protection Agency (EPA) published the final rule for Regional Haze Regulations in the Federal Register on July 1, 1999 (FR Vol. 64, No. 126, and pgs.-35714-35774). The Regional Haze Rule was authorized under Section 169A of the Clean Air Act which sets a national goal for visibility as follows: *"prevention of any future, and the remedying of any existing, impairment of visibility in Class I areas which impairment results from manmade air pollution."*

The sources of visibility reducing pollutants and the mechanisms that result in regional visibility degradation span geopolitical boundaries and, therefore, cooperative programs involving groups of states will facilitate the development and implementation of effective responses for mitigating regional haze. Multi-state emissions inventories of the sources of visibility reducing pollutants will be needed as input to regional models that will be used to evaluate current conditions and mitigation strategies.

Because regional haze is a new program that involves several sources and pollutants that have not been included in previous emissions inventory development efforts, there are significant uncertainties in current estimates of some of the important emissions. The only emissions database that is available on the appropriate regional scales to address regional haze is the National Emissions Inventory (NEI) formerly known as the National Emissions Trends (NET) inventory. The NEI inventory is prepared by the U. S. EPA and represents a consistent national compilation of all sources of pollutants based on a common set of methods. For this project, the latest complete and quality assured year of record represented in the NEI inventory is for 1996. This 1996 NEI inventory (version 3.12) was used as the basis for all of the analyses of emission estimates in this effort.

The first objective of this paper is to present a concise summary of what is known about the sources of visibility reducing pollutants in the eastern part of the United States. This discussion explains why a new inventory development effort is needed to support regional haze studies. Specifically, the paper discusses the sources of primary fine particulate matter and gaseous precursors to secondary fine particulate matter that have not been of major significance in previous air quality management programs.

The second objective of this paper is to describe the methods that have been used to create the 1996 NEI inventory and to present a summary of the emissions estimates in the 1996 NEI inventory for the different regions included in this study. The weaknesses of those methods for use in regional haze studies are summarized. The implications of those weaknesses for future efforts by states and state organizations to implement projects to improve the regional inventories for haze applications are also discussed.

The third objective of the report is to present a list of activities that could be initiated to improve inventories for regional haze assessments. Several of these activities are being addressed through national programs by the U. S. EPA and other cooperative projects. A simple set of criteria have been applied to set priorities among those activities that can be implemented by State/Tribal/Local agencies in the next five years to supplement those national efforts.

Finally, the fourth objective of this paper is to develop a list of specific activities, in the form of a work plan, that can be used by the MANE-VU and VISTAS states to guide the implementation of high priority tasks that will provide the greatest benefits to improving the regional inventories for haze applications over the next five years.

Multi-State Regions Represented In The Emissions Summaries

Summaries of the principal sources of primary fine particulate and the gaseous precursors to secondary fine particulate are presented for the Eastern U.S. and for groups of Eastern States that have formed cooperative associations. The important sources and the immediate inventory development needs are briefly discussed for each group of states. The states included in each group discussed in this report are listed below.

The NESCAUM Region includes Maine, Vermont, New Hampshire, Massachusetts, Rhode Island, Connecticut, New York, and New Jersey.

The MARAMA region includes New Jersey, Pennsylvania, Delaware, Maryland, West Virginia, Virginia, and North Carolina, as well as the District of Columbia.

The NESCAUM and MARAMA states together (except for North Carolina, Virginia, and West Virginia comprise MANE-VU states.

The VISTAS region includes Kentucky, Tennessee, North Carolina, South Carolina, Georgia, Florida, Alabama, Mississippi, Virginia and West Virginia.

VISIBILITY SOURCES IN THE EASTERN UNITED STATES

The nature of regional haze problems requires emissions inventories of all of the important haze pollutants that extend over multi-state regions. It is desirable to use an inventory database that is based on consistent methods to ensure that all of the important source categories are included and that the emissions estimates are comparable. Currently, the only inventory that meets these requirements is the National Emissions Inventory (NEI, formerly known as the National Emissions Trends, NET inventory.)

All of these discussions concerning existing emissions information for regional haze planning and the methods used to develop those estimates are based on the 1996 NEI Version 3.12. That version of NEI was downloaded from the EPA Internet URL: http://www.epa.gov/ttn/chief/net/index.html, on September 29, 2000. This version of the NEI includes some emissions estimates that were submitted by state agencies for selected source categories and pollutants. $PM_{2.5}$ and NH_3 estimates were prepared by EPA. The base-year 1996 was chosen as the year of record for this analysis because that year represents the most recent year of complete data that had been subject to update and revision as the result of a significant quality assurance review effort.

This discussion focuses on those source categories that represent the major contributions to primary fine particulate emissions and the gaseous precursors to secondary sulfate, and nitrate fine particulate in the Eastern United States. The final report also presents information on the sources of precursors to the organic component of fine particulate matter.[1]

Emission summaries for primary fine particulate matter ($PM_{2.5}$), PM_{10}, and the principal precursors of sulfate and nitrate secondary fine particulate matter (SO_2, NO_x, and NH_3) are summarized below to provide an overview of the types of activities that are important sources of emissions in the east. Bar charts representing the distribution of different groups of sources for each pollutant are presented in Figures 1 through 4 for $PM_{2.5}$, SO_2, NO_x, and NH_3, respectively. Tables with emissions summary data for each individual State in the study area are included in Appendix A to the final report.[1]

These emissions estimates can be used to identify the source category groupings that are likely to be the most significant in terms of contributing to regional haze problems. Since the inventory development process has just begun for some of the important pollutants and source categories, there is uncertainty in many of these estimates. The basic weaknesses in the emissions estimation methodologies used to generate the emissions information are briefly discussed. These weaknesses are addressed in more detail in the final report.

Figure 1 summarizes the primary sources of fine particulate matter (particulate matter with mean aerodynamic diameter of 2.5 μm or less). Fugitive dust sources dominate the emissions magnitude and the other primary sources are all fuel combustion sources. The other combustion category is dominated by open biomass burning, which includes burning of construction and land clearing debris, yard waste, agricultural field residue, and other open burning activities. The dominance of fugitive dust is misleading in terms of its contribution to the regional haze problem. The emissions rates are estimated by measuring the horizontal mass flux immediately downwind from known sources of dust (e.g., construction sites, field tilling, etc.). Analyses of exposed filter media from ambient monitoring stations at Class I areas suggests that dust is a much smaller contribution to the regional haze problem. It has been suggested that most of the dust mass is deposited near the source through gravitational settling, aggregation, and deposition on nearby flora and/or buildings. Much more needs to be known about the behavior of the dust sources and their importance in regional haze problems in the East. The significant differences in total mass emissions between the Northeast and Southeast states implies that understanding sources of dust is an important issue for those states included in the VISTAS region.

Throughout the Eastern States, sulfate is a dominant component of fine particulate mass. This is particularly true for data collected at non-urban sampling locations typical of the Class I areas. While the contribution of carbonaceous matter, resulting primarily from mobile sources, is larger in most Eastern urban locations, sulfate remains a major component of the total mass in all areas in the East. Sulfur dioxide (SO_2) is the principal precursor to sulfate aerosol. Figure 2 presents the major contributions to sulfur dioxide emissions in the East. Electricity generation is the dominant source of SO_2 and the larger contribution from the Southeastern states results from increased use of coal as a fuel for electric generating facilities relative to the Northeast.

Figure 1. Emissions of $PM_{2.5}$ by major source category groupings

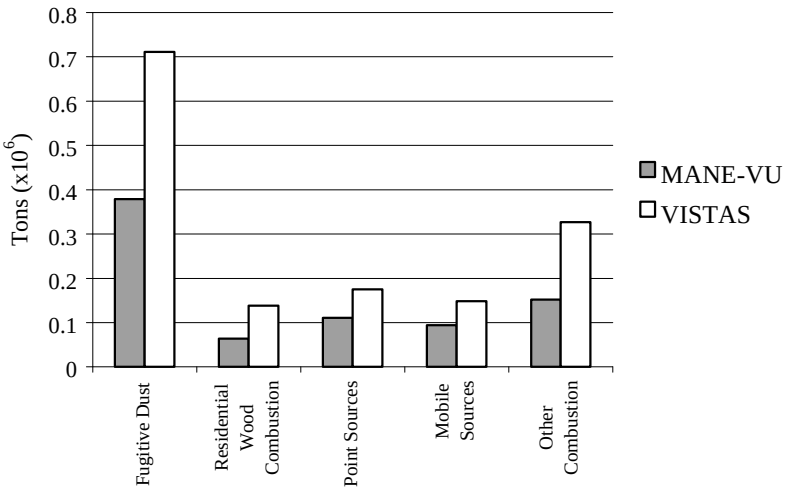


Figure 2. Emissions of SO_2 by major source category groupings

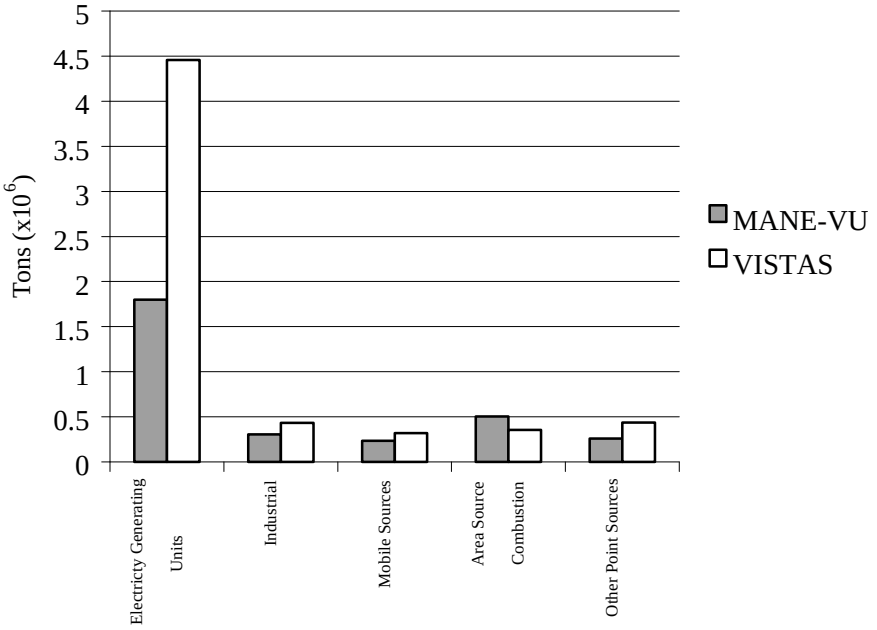
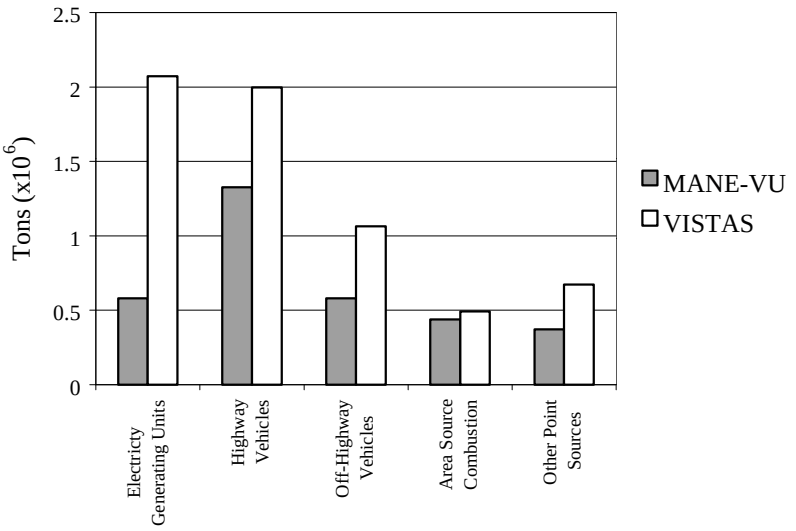


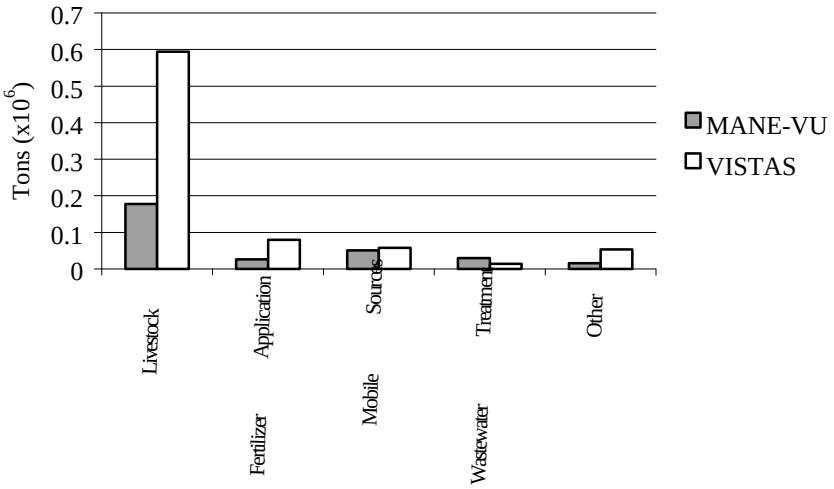
Figure 3 presents the distribution of NO_x emissions in the Eastern United States by major source category grouping. NO_x is an important precursor to nitrate particulate matter. While nitrate represents a small component of the overall fine particulate matter in the East, there is some evidence that suggests that nitrate may become more important as the precursors to sulfate particulate are reduced. This shift in the potential for nitrate formation may result from a change in the competitive reaction pathways through which ammonia is removed from the atmosphere.[2] While electric utilities are a major contributor to NO_x emissions, highway mobile sources make up a significant component of the NO_x emissions throughout the East. A significant amount of the electric generating capacity in the Northeast results from hydroelectric and nuclear facilities, which reduces the relative magnitude of both SO_2 and NO_x from electric utilities in the Northeast relative to the Southeast, which relies more heavily on coal and other fossil fuels for electricity generation.

Figure 3. Emissions of NO_x by major source category groupings



Finally, Figure 4 presents the contributions of NH_3 in the East by major source category. There is a great deal of uncertainty associated estimating NH_3 emissions. The dominant source through the East is livestock operations, including the production of dairy cattle, and poultry in both the Northeast and Southeast. Beef cattle and hog production categories are also contribute large emissions totals in the Southeast. Uncertainties in NH_3 emissions estimates result from limited information on the numbers of animals produced, the timing of when animals are in full production and in the emission factors that are available. Most of the emission factors recommended for use in the NEI are based on research that has been conducted in Europe. Operational factors, such as the type of feed used, the animal containment systems used, the type of waste management systems used, and meteorological conditions can all affect the rate of NH_3 production and volatilization. Clearly, more research is needed to develop accurate methods for estimating NH_3 emissions in the Eastern United States.

Figure 4. Emissions of NH_3 by major source category groupings



ESTIMATION METHODOLOGIES AND RESEARCH PRIORITIES

The final report describes the methods used to prepare the estimates of the emissions of haze pollutants and precursors summarized above. The critical issues are related to the nature of these pollutants and their relative perceived importance in previous air quality management planning. State/Tribal/Local agencies in the East have gained significant understanding of the major sources that contribute to ozone and PM-10 nonattainment issues. While many of the sources of haze pollutants and precursors overlap with the source categories of interest in these previous programs, some of the characteristics of these sources that are important in regional haze planning were only of secondary concern in ozone and PM-10 planning. Other categories and in the case of NH_3, the pollutant itself, have not been important in these previous programs. Some of the important issues related to fine particulate and haze precursor emissions are discussed below.

PM-10 nonattainment is largely a local problem. Fugitive dust sources contribute most of the mass of PM-10. Since mass is a function of the cube of particle radius, the smaller particles that make up the fine particulate matter fraction have been of limited concern. Control of the large particles in the course size range (2.5 µm to 10.0 µm) offers a sufficient strategy to reduce PM-10 mass loading to levels that comply with the PM-10 NAAQS in most circumstances. The smaller particles that are available for long range transport and, therefore, participate in visibility reduction at Class I areas have simply never been a major priority in comprehensive PM-10 planning programs.

The discrepancy between emission rates of fine particulate matter from fugitive sources using the horizontal flux measurement approach and concentrations measured on filters in remote Class I areas has been discussed earlier. The mechanisms for removal of fine particle mass and the factors that control the amount of fine particulate mass that is transported need to be understood in terms of their relative contribution to visibility degradation. A significant body of work has been developed by the Western Regional Air Partnership (WRAP) on the sources of fugitive dust and the role of fugitive emissions of fine particulate matter in visibility

degradation.[3] This report and other information on fugitive dust emissions is available at the Research and Development Forum Web Page at URL: http://www.wrapair.org/commindex.htm These sources are likely to be of less concern in the Eastern United States, relative to the west, due to the significant differences in soil types, land cover and soil moisture content. Appropriate emissions estimates from these sources are needed for two reasons. These sources must be understood to accurately simulate the total particle loading that causes haze and to develop appropriate control strategies. In addition, when planning efforts related to PM-2.5 nonattainment programs begin these sources may be important contributions to population exposure in communities near fugitive dust sources.

The role of area source combustion processes in fine particulate emissions is also poorly understood. This is particularly true for open burning combustion sources of biomass fuels. With the exception of selected communities where emissions from residential wood combustion are important in PM-10 planning, emissions of fine particulate from biomass combustion sources have been of limited concern in most PM-10 nonattainment areas. These sources may be critically important, however, in understanding the source mix and developing effective control strategies to improve visibility in many parts of the country. Important information can be gained by tracking the two dominant components of organic aerosol, elemental carbon and organic carbon. Elemental carbon, often referred to as black carbon, is essentially soot. The term organic carbon is used to describe other chemically bound forms of carbonaceous aerosol. Some of the organic carbon particulate matter is formed at the source, but some is produced through complex heterogeneous chemistry in the atmosphere. Sources that may be of importance to regional haze planning in the East include land clearing and construction waste debris burning, agricultural field burning, and residential waste burning. In selected areas, particularly some areas in the Southeast, prescribed burning and wildfires are also important sources. Much more needs to be known about the source activities and appropriate emission factors for these types of sources.

Point and mobile combustion sources using fossil fuels are also important in regional haze studies. The major uncertainty associated with fine particulate emissions from such sources is associated with condensable material. Condensable materials are pollutants that are in a gaseous form in the hot flue gas or tail pipe exhaust, but immediately condense to form aerosol particulate when mixed with cooler ambient air. The standard methods for monitoring PM-10 from point sources, Method 5, specifies the use of a heated inlet to limit the amount of gas-to-particle conversion prior to sampling. Therefore, many of the existing emission factors for PM-2.5 do not include the fraction represented by the condensable components. EPA is working to correct this problem, by updating appropriate sections of AP-42. As an example, the condensable fine particulate emission factors for natural gas combustion are about 3 times the emission factor for the filterable component of PM-2.5.[4] Additional work is needed to clearly understand the total PM-2.5 emission rates and the relative amounts of elemental and organic carbon from most combustion sources.

In the Eastern United States sulfate aerosol is the dominant form of fine particulate matter that affects visibility. Although the principle sources of SO_2 and NO_x have been well studied, uncertainties remain, particularly for area sources. Overall the contribution of near surface area sources is small compared to large point combustion sources and in the case of NO_x mobile

sources. Smaller individual sources that make up the area source categories may be important in haze formation processes. These emissions may contribute the sulfur and nitrogen that are available to participate in the chemical conversion processes that form aerosol. These processes are associated with NH_3 and are moderated by relative humidity, both of which are major features of the near surface layers of the atmosphere in the East. The chemical processes that form acid aerosol, and the various intermediate aerosol species that ultimately result in ammonium sulfate and ammonium nitrate are not well known. Specifically, the role of near surface sources, the mechanics and rates of competitive reactions that form ammonium sulfate and ammonium nitrate need further study.

There is a great deal of uncertainty associated with emissions estimates of NH_3 in the Eastern United States. Current estimates are based on incomplete information on both the activity levels and emission factors used to represent the major sources of NH_3. In addition, many of the minor sources are ignored completely in current national level inventories. MARAMA and NESCAUM have completed a project to develop an emissions estimation model for NH_3 that incorporated features that make it easy to update both activity data and emission factors for most important sources of NH_3 as improved information is made available. The NH_3 emissions model and user guides can be downloaded from the Internet, URL: (http://www.envinst.cmu.edu/nh3/). Clearly, there is a significant need to improve the methods and tools for estimating ammonia emissions.

Finally, the study addressed the issue of temporal, spatial and species resolution of emissions estimates for application to regional modeling. The current national and regional scale emissions inventories are developed at annual resolution (emissions are expressed in units of tons/year). Regional haze occurs throughout the year, and the sources that contribute to haze can vary by season. Therefore, visibility degradation at Class I areas can be caused by a different mix of sources in different seasons. It is important, therefore, to include information that can be used to accurately partition annual emissions to seasons when information is collected to describe the base activity rates of the primary sources.

Area sources are estimated at the county-level in national inventories. These emissions must be further spatially distributed to conform to the selected grid spacing and dimensions of the applicable air quality model. This step is accomplished by use of spatial distribution surrogates. For example, sources that are associated with human activities are distributed by use of a population surrogate distribution file. There are a limited number of these surrogate factors and some sources must be assigned factors that may not accurately represent their distribution. Furthermore, spatial patterns are not static in time. The planning horizon for the regional haze program extends 60 years into the future. Spatial patterns of agricultural land, forested land, and population can change over periods of five to ten years in rapidly growing communities. Therefore, there are needs related to improving spatial distribution surrogates for current analyses and for developing spatial distribution surrogates to represent expected future conditions. The Emissions Inventory Improvement Program (EIIP) has initiated a project to collect and assemble information to prepare future spatial allocation factors that will cover, at a minimum, major growing urban areas.

Currently, emissions of particulate matter and volatile organic compounds (VOC) are reported as an aggregate mass emission rate. Each of these pollutants represents the total of hundreds of specific chemical components. Each of these components interacts in the chemical and physical processes that control the distribution and transformation of secondary pollutants at different rates and in different ways. There are few high-quality, publicly available speciation profiles that were developed specifically for fine particulate matter sources. The existing VOC profiles were developed primary for application to ozone formation systems and may not include complete information on some of the heavier organic compounds that participate in organic aerosol formation. Further work is needed to expand both the PM and VOC speciation databases for application to regional haze problems.

DEVELOPMENT OF A FIVE-YEAR WORK PLAN

MARAMA, in cooperation with MANE-VU, evaluated the current state of knowledge concerning the important emissions sources of haze pollutants and developed a work plan to guide research and development efforts over the five period between 2001 and 2005.[5] This work plan was based on the recommendations presented in the emissions inventory assessment report discussed earlier.[1] The following list of criteria were used to select the high priority tasks and to group them into a year-by-year schedule:

- The need to spread activities over a multi-year period,
- The availability of supporting information,
- The priority of various source categories to regional haze planning in the East,
- The anticipated availability of new methods and approaches under development by other RPOs and EPA.

The following high priority needs have been segregated as activities that can be best handled at the national-level with EPA support:

- An accepted source measurement method that is capable of measuring both filterable and condensable components of fine particulate matter,
- An understanding of the mechanisms that alter or decrease the mass of primary fine particulate matter from fugitive dust sources, and how those emissions should be treated in regional haze modeling scenarios,
- Improved understanding of the chemical mechanisms that control gas-to-particle conversion processes, and implementation of those mechanisms into model compatible modules,
- Improved emission factors for most major ammonia sources,
- A better understanding of the sources and sinks of ammonia and how those sources and sinks interact under different meteorological conditions, and
- Improved speciation factors for fine particulate matter and VOC sources.

The recommended projects included in the work plan are listed in Table 1. MARAMA plans to update this work plan annually to reflect new information.

Table 1. Recommended Work Plan Tasks for Regional Haze Planning in the Northeast States

Task Number	Description	Suggested Lead Agency
Tasks for Calendar Year 2001		
1	Review CMU NH_3 inventory for use in planning	States/Tribes
2	Review and correct point source physical data	States/Tribes
3	Identify point sources of condensable matter	States/Tribes with EPA guidance
4	Improved activity for RWC and open burning	MARAMA contractor
5	Develop GIS tools for emissions allocation	MARAMA
6	Identify BART eligible sources	States/Tribes
Tasks for Calendar Year 2002		
7	Collect activity data for area combustion sources	States/Tribes
8	Collect activity data for nonroad mobile sources	States/Tribes
9	Improve activity data for rural highway sources	States/Tribes
10	Process for reporting EC/OC emission splits	EPA with multiregional effort
11	Identify sources of agricultural activity data	EPA, USDA, and States/Tribes
12	Assemble improved emissions estimates for RWC, and open burning	MARAMA contractor
Tasks for Calendar Year 2003		
13	Collect activity data for paved and unpaved roads	States/Tribes with EPA guidance
14	Collect activity data for agricultural sources of NH_3	States/Tribes
15	Identify and assess miscellaneous NH_3 point sources	States/Tribes
16	Compile 2002 regional point source emissions inventory	States/Tribes with MARAMA contractor
Tasks for Calendar Year 2004		
17	Compile 2002 regional area, nonroad and mobile source emissions inventory	States/Tribes with MARAMA contractor
18	Develop activity data for natural sources	EPA with multiregional effort
Tasks for Calendar Year 2005		
19	Assess and address remaining weaknesses in emissions inventory	States/Tribes with MARAMA contractor
20	Assemble and quality assure emission estimates for modeling and source apportionment studies	States/Tribes with MARAMA contractor

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