VISTAS 2002 Emissions Inventory Development

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ABSTRACT

The organization Visibility Improvement - State and Tribal Association of the Southeast (VISTAS) is a collaborative effort of the ten southeastern states and eligible tribes to conduct technical analyses and planning activities addressing regional haze and related air quality issues. VISTAS is developing a 2002 base year emissions inventory in cooperation with the southeastern state and local air quality agencies and with technical support from E.H. Pechan and Associates, Inc. and MACTEC Engineering and Consulting, Inc. The 2002 emissions inventory is intended to support emissions and photochemical modeling for regional haze and fine particulate matter. The inventory will include primary and precursor emissions for fine particulate matter. Particular attention will be paid to developing speciation factors to describe primary sources of organic carbon, elemental carbon, crustal components, and trace elements. Seasonal, weekday/weekend, and diurnal adjustment factors will be developed as inputs to the emission model. VISTAS is prioritizing emissions inventory improvements based on composition of fine particulate matter in the southeastern United States and uncertainty in emissions inventories for key components. VISTAS will improve activity factors for agricultural sources of ammonia and fire and will address uncertainties in emissions factors for organic carbon and elemental carbon from anthropogenic and biogenic sources, including combustion of coal, gasoline, diesel, wood, and vegetative emissions.

INTRODUCTION

The Southeastern States Air Resource Managers, Inc. (SESARM) has been designated by the United States Environmental Protection Agency (EPA) as the entity responsible for regional haze planning for the eight SESARM states (Alabama, Florida, Georgia, Kentucky, Mississippi, North Carolina, South Carolina and Tennessee) plus Virginia, West Virginia, and the Eastern Band of Cherokee Indians. These parties are collaborating through the organization known as Visibility Improvement - State and Tribal Association of the Southeast (VISTAS) in the technical analyses and planning activities associated with visibility and related regional air quality issues. VISTAS analyses will support the states in their responsibility to develop, adopt, and implement

their State Implementation Plans (SIPs) for regional haze. The VISTAS states and the 18 Class I areas (national parks greater than 6,000 acres and wilderness areas greater than 5,000 acres) in the VISTAS region are illustrated in Figure 1.

Figure 1. VISTAS region and Class I areas.

VISTAS is developing a 2002 emissions inventory to support atmospheric modeling of fine particulate matter (fine particles less than 2.5 microns in diameter, $PM_{2.5}$) and visibility. The draft 2002 inventory will be based on the 1999 National Emissions Inventory and updated with data from the VISTAS state and local air quality agencies. This paper summarizes the emissions trends in the VISTAS states and work underway to improve current inventories.

EMISSIONS INVENTORY

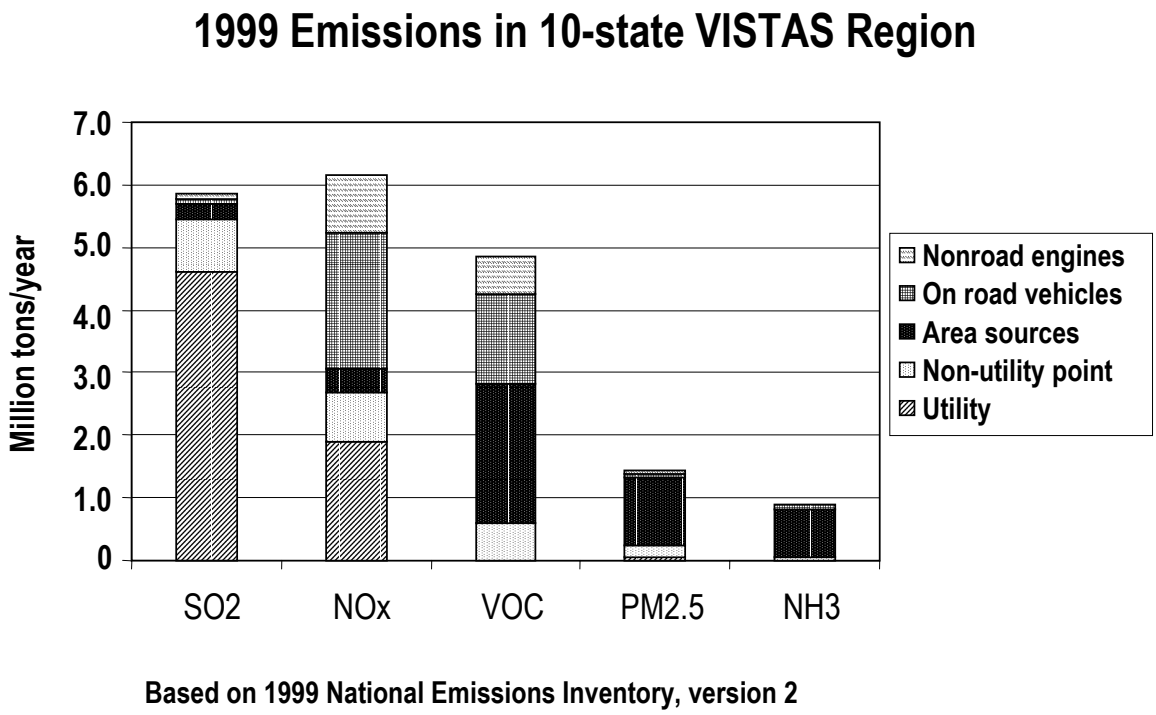
VISTAS is working with state and local air quality regulatory agencies and two consulting firms, MACTEC, Inc. (MACTEC) and E. H. Pechan and Associates, Inc. (Pechan) to develop a 2002 emissions inventory appropriate for VISTAS emissions and air quality modeling. The 1999 National Emission Inventory Version 2 (NEIv2)[1] is the starting point for the 2002 inventory. The 2002 inventory will include:

- All primary and precursor emissions necessary to accurately model $PM_{2.5}$, specifically primary $PM_{2.5}$ and particles with diameter less than 10 microns

(PM_{10}), sulfur dioxide (SO_2), nitrogen oxides (NO_x), volatile organic compounds (VOC), ammonia (NH_3), and carbon monoxide (CO),
- Speciation factors for primary $PM_{2.5}$ and PM_{10}, including factors for elemental carbon (EC), organic carbon (OC), and crustal elements (e.g. carbon, magnesium, potassium, phosphorus),
- Seasonal, weekday/weekend, and diurnal adjustment factors for each source category.

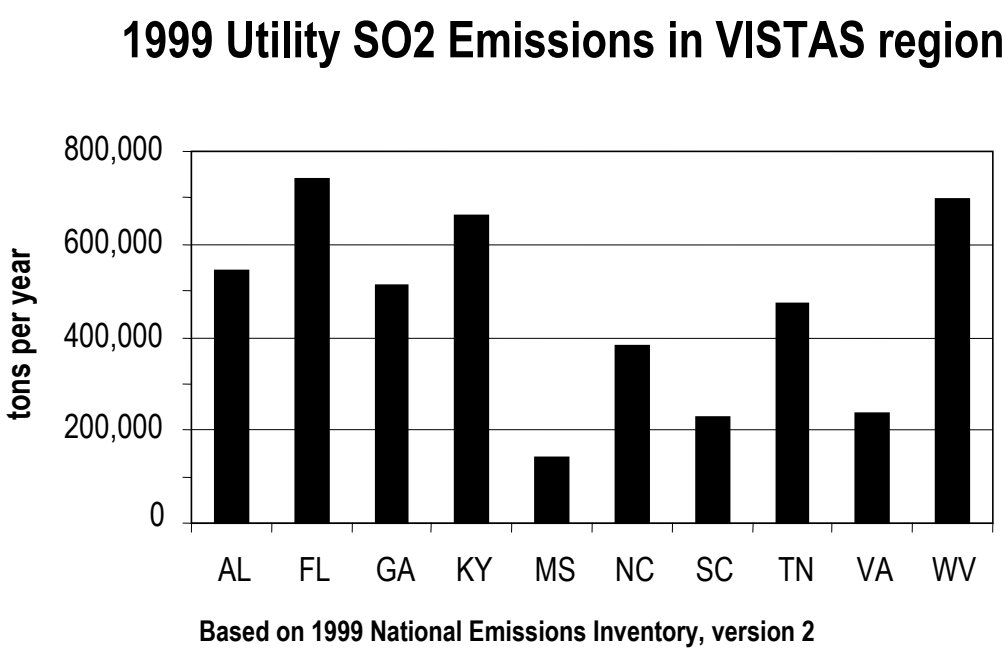
Contributions of the major source sectors to inventories for SO_2, NO_x, VOC, primary $PM_{2.5}$, and NH_3 in the ten VISTAS states are illustrated in Figure 2.

Figure 2. 1999 Emissions in VISTAS states.



Utility coal combustion is the most important source of SO_2 in the VISTAS states, comprising 70% of total SO_2 emissions in the region in 1999. Other contributions are from utility oil combustion, industrial point and area sources and nonroad engines. Florida, West Virginia, and Kentucky have the largest utility SO_2 emissions in the VISTAS region (Figure 3).

Figure 3. 1999 Sulfur Dioxide (SO_2) Emissions in the VISTAS states.



Nitrogen oxide (NO_x) emissions are primarily from utility coal combustion, diesel highway vehicles, gasoline highway vehicles, and nonroad engines. Collectively these sources account for over 75% of total NO_x emissions in the VISTAS region. Other types of fuel combustion (e.g. utility natural gas, industrial, residential) account for an additional 14% of the total NO_x emissions. Florida, Georgia, and North Carolina have the highest NO_x emissions in the VISTAS region. These emissions trends follow state population trends.

Highway vehicles, area sources, and nonroad engines are the major sources of anthropogenic VOCs. Burning categories (wildfires, prescribed burning, open burning, residential wood combustion) account for 14% of the anthropogenic VOC inventory. Biogenic VOC emissions, such as those from vegetation, are not accounted in the NEI but are calculated in the atmospheric model.

Paved and unpaved roads, agricultural crop/livestock activities, and construction activities comprise 40% of the primary $PM_{2.5}$ inventory. Burning activities (wildfires, prescribed burning, open burning, residential wood combustion) comprise another 12-24% of the $PM_{2.5}$ inventory. These sources also are important for organic carbon and elemental carbon components of $PM_{2.5}$.

Area sources are the predominant contributors to the NH_3 inventory. Livestock operations, fertilizer application, and wastewater treatment processes represent over 85 % of the total NH_3 inventory for the VISTAS region. Agricultural chemical manufacturing is the primary ammonia point source category in the NEI.

VISTAS Emissions Inventory Updates

During fall 2002, state and local air quality agencies provided the most recent inventory information (1999, 2000, 2001, or in some cases 2002) for each major source sector [2,3]. In particular, updated information was provided on vehicle miles traveled (by road type and vehicle class), fuel use, and local control programs. Missing data in the updated point and area source inventories (e.g. $PM_{2.5}$, NH_3) were filled from the 1999 NEIv2, using appropriate growth factors. Growth factors from the Bureau of Census and the Economic Growth Analysis System will be used to project emissions for 2002. Inventory objectives, methods, and results will be documented in a quality assurance plan, consistent with EPA's Guidance for Developing an Emissions Inventory[4].

For point sources, working with state and local agencies, selected sources will be surveyed to validate location, stack parameters, and seasonal emissions characteristics. For several large point sources, discrepancies in the NH_3 emissions between the Toxics Release Inventory and the NEI will be reviewed and reconciled. Continuous emissions monitoring data for 2002 will be incorporated as available for specific point sources. The Carnegie Mellon University (CMU) NH_3 model will be applied to define NH_3 emissions from area sources.

MOBILE6 will be used to generate emissions for on-road vehicles. The NONROAD model (spring 2003 release) will be applied for appropriate source sectors. Fuel data updated for on-road vehicles will be applied as appropriate for the nonroad sectors. Ammonia emissions are not provided in the NONROAD model but will be estimated using fuel data for these sectors. Emissions from aircraft, commercial marine vessels, and locomotives will be developed using activity factors from the 1999 NEIv2 and grown to 2002. Specific data for airport landings and takeoffs will be applied where provided by state and local agencies.

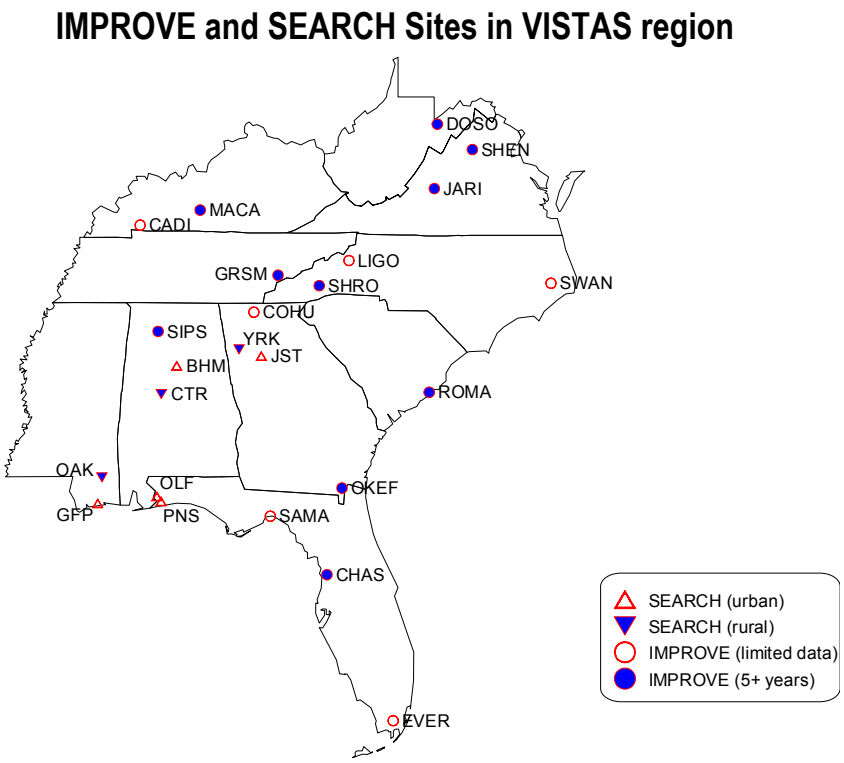
The first draft 2002 inventory will be provided to the state and local agencies for review in June 2003. State review comments and available new activity and emissions data will be incorporated in the fall of 2003. The 2002 inventory will be projected to the appropriate period for the emissions and air quality modeling. Inputs for VISTAS emissions modeling are to be delivered by the beginning of 2004. Growth projections for future year emissions inventories and control strategies will begin during the fall of 2003.

VISTAS Emissions Inventory Priorities Based on Air Quality Monitoring Data

VISTAS is using air quality monitoring data in the VISTAS region to assist in prioritizing emission inventory improvement needs. VISTAS reviewed speciated PM2.5 data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network[5] for the period 1998 to 2001 and from the Southeast Aerosol Research Characterization Study (SEARCH) network[6] for the period 1999 to 2001. The 15

IMPROVE sites represent the range across the 18 Class I areas in the VISTAS region from Southern Appalachian Mountains to the Gulf and Atlantic coasts (Figure 4). The SEARCH network includes 4 urban areas (Atlanta, GA; Birmingham, AL; Gulfport, MS; and Pensacola, FL) paired with a nearby rural (or suburban in the case of Pensacola) site for a total of eight sites.

Figure 4. IMPROVE and SEARCH sites in VISTAS region.



The components of $PM_{2.5}$ mass and their contribution to visibility impairment were evaluated for days with the 20% best and 20% poorest visibility (Figures 5 and 6) [7]. Visibility is measured as the scattering or absorption of light by particles and gases in the atmosphere (collectively called light extinction). Visibility can be measured directly or calculated using the mass and extinction efficiencies of $PM_{2.5}$ components and relative humidity of the atmosphere.

Sulfate fine particles are the dominant contributors (60-80%) to fine particle mass and light extinction on the 20% poorest visibility days at all IMPROVE and SEARCH sites in all quarters of the year. Sulfate levels are typically higher at the Southern Appalachian IMPROVE and northern SEARCH sites than at the southern and coastal sites in either network. Sulfate levels are typically highest in the summer months. Improved information on seasonal variation in SO_2 emissions and improved precision of point, area, and nonroad inventories for SO2 are recommended.

Figure 5. Light Extinction on Days with 20% Poorest Visibility at IMPROVE monitoring sites in 1998 to 2001.

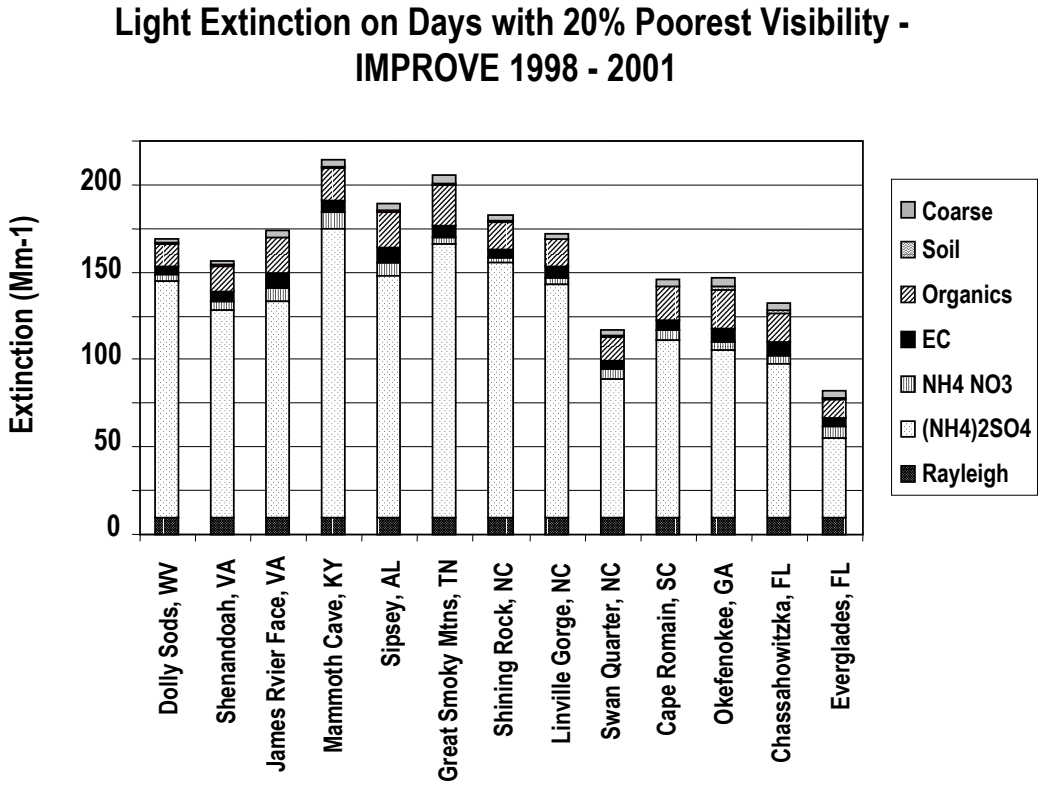
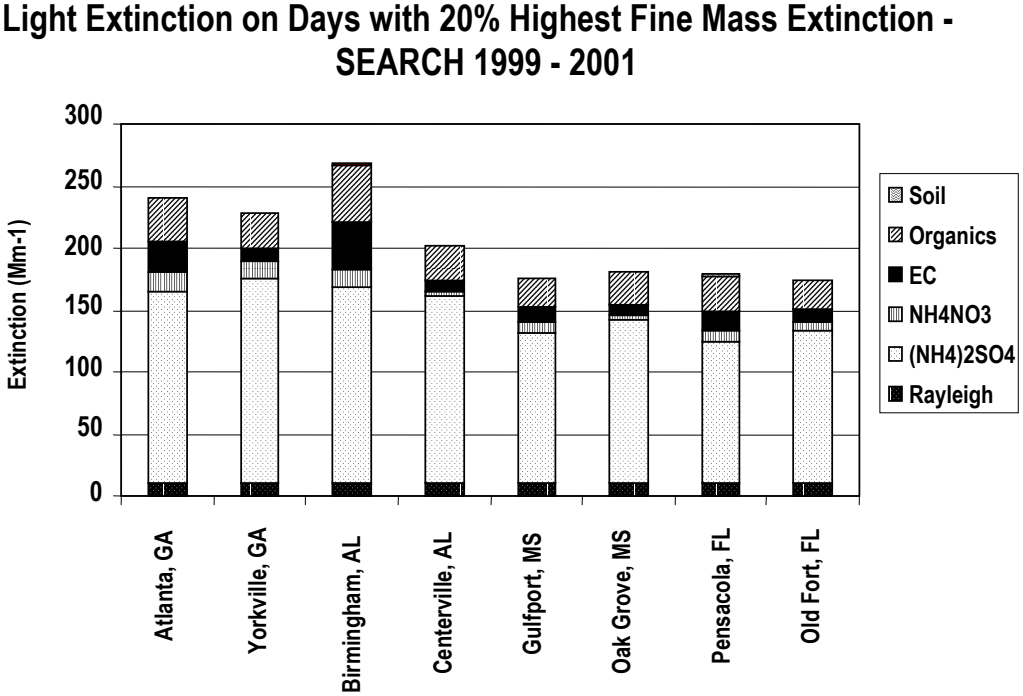


Figure 6. Light extinction due to fine mass on days with 20% poorest visibility at SEARCH sites in 1999-2001.



Sulfate fine particles occur as either ammonium sulfate, $(NH_4)_2SO_4$; ammonium bisulfate, NH_4HSO_4; or sulfuric acid, H_2SO_4. In the IMPROVE network, ammonium is not routinely measured. Measured sulfate is assumed to be fully neutralized by ammonium and to occur as $(NH_4)_2SO_4$. Measurements of ammonium at 3 IMPROVE sites and at the SEARCH sites indicate that sulfate is not fully neutralized by ammonium, particularly during the summer months. Ammonium preferentially neutralizes sulfate particles before reacting with nitric acid to form ammonium nitrate, NH_3NO_3. NH_3NO_3 levels are typically very low at the IMPROVE sites.

Modeling analyses[8] suggest that, in the southeastern US, NH_3NO_3 formation is limited by ammonium availability in the atmosphere. NH_3NO_3 is most likely to be formed early in the morning when relative humidity is very high. Because NH_3NO_3 volatilizes at warmer temperature, nitrate particles are higher in the fall and winter quarters than in the warmer months. NH_3NO_3 levels are higher in the SEARCH urban areas where nitrogen oxide emissions are more concentrated. These results indicate that accurate estimates of ammonia emissions are needed to accurately model the degree of neutralization of $(NH_4)_2SO_4$ and levels of NH_3NO_3 in the atmosphere. Accurate estimates of the seasonal variation in ammonia emissions and spatial variation in both ammonia and nitrogen oxide emissions are needed.

Organic carbon is the second largest contributor (5-20%) to fine particle mass and light extinction on the 20% poorest visibility days in both networks. Organic carbon levels are higher in the urban SEARCH sites than at the rural SEARCH sites and the IMPROVE sites, suggesting a mix of regional and local sources in the urban areas. Improving inventories of primary organic carbon is a high priority for VISTAS.

The contribution of organic carbon mass to light extinction is relatively constant across the quarters. The IMPROVE and SEARCH measurements of organic carbon do not allow separation of primary from secondary carbon sources nor anthropogenic from biogenic sources. Chemical mass balance analyses for the SEARCH sites[9] suggest that primary organic carbon is predominantly from diesel, gasoline, and wood burning sources. Wood burning has the highest contribution to total organic carbon in the fall and winter months. Differences between total organic carbon and primary carbon can be attributed to secondary organic carbon, primarily of biogenic origin. This component of total organic carbon is highest in the spring and summer months and consistent with emissions from vegetation. Carbon-14 analyses can separate carbon of recent origin (e.g. wood burning and vegetative emissions) from carbon of fossil origin (e.g. coal, diesel, and gasoline). Carbon-14 analyses for the SEARCH sites[10] indicate that in the fall and winter total organic carbon is equally divided between fossil and recent carbon while in the spring and summer, recent carbon has a higher contribution to total organic carbon. Atmospheric modeling of biogenic emissions in the southeastern U.S.[8] indicates that biogenic emissions of volatile organic carbon and organic aerosols are significantly greater than anthropogenic sources. Improvements in understanding of seasonal variation in emissions of primary organic carbon and volatile organic carbon are recommended.

Elemental carbon generally contributes less than 10% to light extinction at the IMPROVE and 5-20% at the SEARCH sites. Comparatively higher levels of elemental carbon in the urban SEARCH sites indicate a mix of local urban and regional sources. Improvements to speciation of primary $PM_{2.5}$ for elemental carbon are a priority for VISTAS inventory efforts.

Episodes of elevated organic carbon, elemental carbon, and non-soil potassium indicate episodes when fire plumes impacted the monitoring sites[11]. Some of the days with the highest light extinction occurred during the fall of 2000 and the fall of 2001 when forest fires contributed to poor visibility at several sites across the VISTAS region. Better inventories of fire occurrence and fuel loading are needed to better estimate impact of fire on visibility and $PM_{2.5}$ levels.

Soil generally contributes 0-3 % of the fine mass extinction and 5-20% of the fine particle mass at the SEARCH and IMPROVE sites. Soil has a higher percentage contribution to fine mass and light extinction at Chassahowitzka and Everglades in Florida than at the other IMPROVE and SEARCH sites. While trajectory analyses and chemical composition of soil constituents indicated that episodes influenced by Saharan dust do occur in the Southeast[11], these appear to be relatively infrequent. Where coarse mass is measured, coarse mass generally contributes less than 5% to total extinction. As a result VISTAS has placed less emphasis on refining inventory for soil and coarse mass.

In summary, these results suggest the following priorities for VISTAS inventory improvements:

- Speciation of $PM_{2.5}$ for organic carbon and elemental carbon, including seasonal variation in emissions
- Accurate estimates of ammonia emissions, seasonal variation, and spatial allocation in ammonia emissions
- Frequency and extent of prescribed fires and wild fires
- Precision in sulfur dioxide emissions

VISTAS Emissions Inventory Improvements

Several inventory improvement tasks are underway in 2003.

<u>Organic Carbon and Elemental Carbon</u>

The SPECIATE data base is being applied to the primary $PM_{2.5}$ inventory in the 1999 NEIv2 to define the major source sectors that contribute to organic carbon and elemental carbon in primary $PM_{2.5}$ in the VISTAS states. This information will be compared to the chemical mass balance results that identify the relative contributions of different emissions types to measured organic carbon. Comparison of inventory and monitoring results will provide insight into next steps for inventory improvements. Under contract to EPA, MACTEC is updating the SPECIATE data base with the most recently available

source sector emissions factors. The updated SPECIATE data base is expected by fall 2003.

Ammonia emissions

The Carnegie Mellon University (CMU) ammonia inventory model will be used to define area source ammonia emissions for the 2002 VISTAS inventory. The VISTAS state air quality agencies have been asked to research available data on livestock operations and activity levels in their state. These data will be used to update activity data available from the Department of Agriculture. Under contract to the Midwest Regional Planning Organization, Sonoma Technology, Inc.[12] has recommended improvements to the emissions factors used in the CMU model. MACTEC will review these recommendations and also recommendations for ammonia emissions factors from a joint study by EPA and North Carolina[13]. Information on seasonal variation in emissions is also being sought.

Fire Emissions

State forestry agencies are being asked to provide electronic records of forest fire and prescribed fire incidence in 2002 to define a 2002 fire inventory. Availability of electronic records varies among the states. Fire records from additional years are also being requested to develop a "typical" year fire inventory for purposes of future year modeling. Information on agricultural burning and open burning will also be sought.

Sulfur Dioxide Emissions

Continuous Emissions Monitoring data will provide detailed emissions information for large point sources. In addition, MACTEC will survey large point sources to assist in the evaluation of emissions and facility location data. Additional improvements for activity factors for area sources and nonroad engines will be considered based on results of the draft 2002 inventory.

Cooperative Inventory Efforts

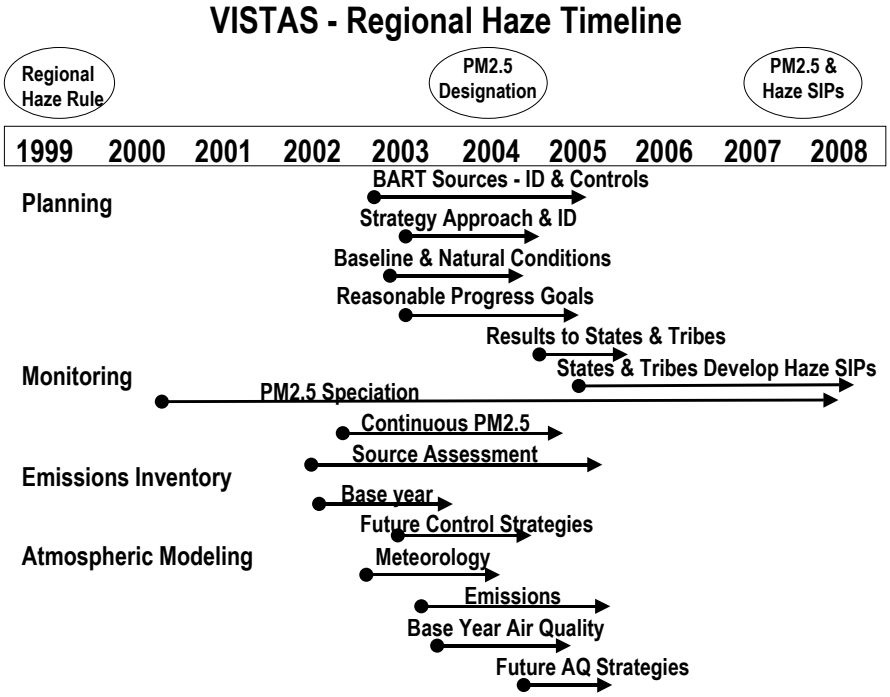
VISTAS is participating with EPA and the other regional planning organizations in discussions of inventory improvement needs. Improvements in emissions factors from projects by EPA, other regional planning organizations, and state-EPA cooperative emissions inventory improvement projects that are available by the end of 2003 will be incorporated into the VISTAS draft 2002 inventory.

VISTAS Emissions Inventory Application for Atmospheric Modeling

VISTAS is testing the MM5 meteorological model MM5, the SMOKE emission model, and the CMAQ air quality model for three episodes in 1999, 2001, and 2002. Sensitivity

and performance of alternative model configurations will be tested during 2003 (Figure 7). The 1999 NEIv2 will be used as the basis for these model sensitivity tests. Annual modeling of PM2.5 is scheduled to begin in January 2004. The 2002 draft inventory will be grown as appropriate to support the annual modeling. Inventory projections for future years and for emissions control strategies will begin in 2004 and will be ready for modeling in late 2004 and 2005. These activities will support state regulatory planning and development of state implementation plans for regional haze.

Figure 7. VISTAS timeline for technical analyses supporting regional haze**.**



CONCLUSIONS

VISTAS is currently developing a draft 2002 emissions inventory based on data provided by the state and local agencies and grown from the 1999 National Emissions Inventory version 2. The draft 2002 inventory will be reviewed by the state and local agencies and VISTAS participants during the summer of 2003 and will be updated in the fall of 2003.

VISTAS priorities for emissions inventory improvements include improvements to speciation of organic and elemental carbon, activity factors and emissions factors for sources of ammonia, fire activity data, and improved precision in sulfur dioxide emissions.

VISTAS is developing the technical tools and capabilities to be ready to evaluate emissions control strategies in 2004 and 2005 and to provide support to state implementation planning for regional haze.

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KEYWORD

Emissions Inventory
PM2.5
VISTAS