

Beyond Sulfate: Maintaining Progress towards Visibility and Health Goals

Non-sulfate Strategy Options for Reducing Fine
Particles in MANE-VU Region

Prepared by
Northeast States for Coordinated Air Use Management (NESCAUM)

December 17, 2012

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NON-SULFATE STRATEGY OPTIONS FOR REDUCING FINE PARTICLES IN MANE-VU REGION

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Executive Summary

In 1999, the U.S. Environmental Protection Agency (USEPA) promulgated the Regional Haze Rule establishing a target year of 2064 to reach natural background visibility conditions in national parks and wilderness areas. A significant portion of degraded visibility in these scenic lands (called “Class I areas”) results from light scattering or absorption by fine particulate matter (PM_{2.5} – particles having a diameter of 2.5 micrometers or less). In addition to visibility impacts, PM_{2.5} poses a significant risk to human health. Inhaled PM_{2.5} can penetrate deep into the lungs, and cause a number of serious health problems, including increased risk of premature death for people with cardiovascular disease.

Fine particulate matter comes either directly from emission sources, or is chemically formed in the air from precursor emissions. Currently in the Mid-Atlantic/Northeast Visibility Union (MANE-VU) region, the pollutant particles making the largest contribution to poor visibility conditions are composed of sulfates. The remaining portion of human-caused visibility degradation is largely attributable to nitrates, organic carbon, and light absorbing carbon.

There has been much progress in reducing sulfate pollution that is responsible for a large portion of poor visibility and total PM_{2.5} mass across the region. Natural background visibility conditions, however, cannot be achieved without also reducing levels of the non-sulfate contributors to visibility impairment, particularly carbon and nitrate particles. These non-sulfate aerosols can also cause localized PM_{2.5} “hot spots” occurring at levels of public health concern. Examples include wood combustion (organic carbon) and diesel engine exhaust (black carbon or “soot”).

In the MANE-VU region, visibility has been improving over the years at federally protected parks and wilderness areas. Improving trends are also occurring in MANE-VU urban areas where monitored annual PM_{2.5} mass concentrations are showing significant declines. These can be attributed to measures initiated under the federal Clean Air Act and through regional coordination efforts among the Mid-Atlantic and Northeast states.

In evaluating future opportunities for continuing progress that go beyond sulfate pollution, this report identifies the current largest source sectors of non-sulfate PM_{2.5} pollution in the MANE-VU region. Of the largest sectors, mobile sources (on- and off-highway) stand out as principal emitters of pollutants contributing to non-sulfate aerosols.

A survey of recent scientific studies provides additional insight into non-sulfate control strategies. Of particular note from the science literature is the role of human-related (anthropogenic) emissions in the formation of “natural” (biogenic) secondary organic aerosols. Studies indicate the human-related pollution sources of nitrogen oxides and primary carbonaceous matter help foster formation of secondary organic aerosols in the atmosphere. The organic constituents of these aerosols can come mainly or entirely from natural sources, such as vegetation. It would be mistaken, however, to conclude that these aerosols are uncontrollable for purposes of addressing visibility and PM_{2.5} problems. Research indicates anthropogenic emissions can promote the production of these organic aerosols in the atmosphere.

With mobile sources being an important contributor to formation of many of the non-sulfate aerosols, new measures to reduce mobile source emissions will have an important role in improving visibility and reducing PM_{2.5} concentrations within MANE-VU. Chief among these would be to lower the sulfur content of gasoline used with the existing motor vehicle fleet. This would significantly reduce emissions of aerosol-forming nitrogen oxides and volatile organic compounds by reducing sulfur poisoning effects in the catalytic converters of gasoline vehicles, thus improving their pollution-reducing efficiency. Early retirements of or retrofitting pollution controls on older diesel vehicles and equipment (e.g., marine vessels, highway trucks, locomotives, construction equipment) helps address diesel exhaust pollution. MANE-VU is also looking at a range of control options for nitrogen oxides and volatile organic compounds for a number of source types, such as non-emergency stationary generators, petroleum storage tanks, and consumer products.

Wood combustion is another important particulate source sector that can have localized impacts in confined valleys as well as broader regional impacts during the winter when wood combustion for space heating is at its highest level. National emission standards for wood stoves have not been updated since 1988, while many other increasingly popular wood burning appliances, such as wood pellet heaters and indoor/outdoor wood furnaces, are not currently subject to mandatory national pollution performance standards.

In addition to traditional control measures, there are other options available with multiple benefits that can also reduce aerosol pollution. There are efforts underway within MANE-VU to quantify and incorporate energy efficiency pollution reductions into air quality planning, and to evaluate the benefits of expanded combined heat and power (CHP) applications.

In summary, the multiple constituents and complex atmospheric chemistry of the non-sulfate aerosols point towards multi-pollutant strategies to achieve future public health and visibility goals for the MANE-VU region. This dovetails with the trend in planning efforts across a broad spectrum of air, climate, and energy issues. Future visibility and PM_{2.5} NAAQS planning has the opportunity to take from these efforts, and contribute to them as well.

1. INTRODUCTION

1.1. National Visibility Goal

In the 1977 Amendments to the Clean Air Act, Congress created a national visibility goal for national parks, wilderness areas, and international parks (“Class I areas”) aimed at achieving natural visibility conditions at scenic vistas in these federally-protected lands. The Amendments required the U.S. Environmental Protection Agency (USEPA) to issue rules that would put the country on a path for making “reasonable progress” towards the national goal.¹ In 1999, more than 20 years after the 1977 Amendments, the USEPA issued the Regional Haze Rule² with a target year of 2064 to achieve the natural visibility goal established by Congress in 1977.

Within the region covered by the Mid-Atlantic/Northeast Visibility Union (MANE-VU),³ the federal Class I areas encompassed by Congress’ national visibility goal are;

- Brigantine Wilderness Area, NJ
- Lye Brook Wilderness Area, VT
- Great Gulf Wilderness Area, NH
- Presidential Range-Dry River Wilderness Area, NH
- Acadia National Park, ME
- Moosehorn Wilderness Area, ME
- Roosevelt-Campobello International Park, ME/NB

A significant portion of reduced visibility below natural conditions in these areas results from light scattering or absorption by fine particulate matter (PM_{2.5} – particles having a diameter of 2.5 micrometers or less). PM_{2.5} is emitted either directly by pollution sources, or chemically formed in the air from precursor pollutants. Currently in the MANE-VU region, the pollutant particles making the largest contribution to poor visibility are composed of sulfates. The remaining portion of human-caused visibility degradation is largely attributable to nitrates, organic carbon, and light absorbing carbon.⁴

1.2. National Health and Welfare Standards for Particulate Matter

In addition to visibility impacts, PM_{2.5} poses a significant risk to human health due to its ability to penetrate deep into the lungs and pass into the bloodstream. In the lungs, PM_{2.5} can aggravate asthma symptoms, promote development of chronic

¹ USEPA (U.S. Environmental Protection Agency). (1999). *Fact Sheet: Final Regional Haze Regulations for Protection of Visibility in National Parks and Wilderness Areas*, April 22, 1999.

² 64 Fed. Reg. 35714 (July 1, 1999).

³ The MANE-VU region covers Connecticut, Delaware, District of Columbia, Maine, Maryland, Massachusetts, New Hampshire, New Jersey, New York, Pennsylvania, Rhode Island, and Vermont.

⁴ IMPROVE (Interagency Monitoring for Protected Visual Environments) Report V. (2011). *Spatial and Seasonal Patterns and Temporal Variability of Haze and its Constituents in the United States*. Prepared by Hand, J.L. et al. ISSN 0737-5352-87 (June). Available at <http://vista.cira.colostate.edu/improve/publications/Reports/2011/2011.htm>.

respiratory disease, and reduce overall lung function. In the bloodstream, PM_{2.5} can lead to heartbeat irregularities, heart attacks, and even premature death in people with cardiovascular disease.⁵

PM_{2.5} levels have dropped in the Northeast overall due to reductions in direct PM_{2.5} emissions as well as emissions reductions of precursor pollutants within the Northeast and in upwind regions. Air quality planners expect that if current progress continues, all areas of the Northeast should meet the 2006 PM_{2.5} national ambient air quality standards by 2015 (15 µg/m³ annual, 35 µg/m³ daily). In December 2012, the USEPA further lowered the annual PM_{2.5} health standard to 12 µg/m³ in light of research indicating health impacts occurred at PM_{2.5} concentrations below the 2006 annual standard.⁶ The USEPA projected that all areas outside of seven counties in California would meet the more stringent annual PM_{2.5} health standard by 2020 as a result of federal air quality rules already in place.

1.3. Beyond Sulfate

Even with continued progress in reducing sulfate levels in MANE-VU Class I areas, natural background visibility conditions cannot be achieved without also reducing levels of the non-sulfate contributors to visibility impairment, particularly organic carbon and nitrate particles.⁷ In addition, while progress is also occurring at the regional level towards meeting the national PM_{2.5} health standards, localized PM_{2.5} “hot spots” occur at levels of public health concern due to numerous non-sulfate emission sources, such as wood combustion⁸ (organic carbon) and diesel engine exhaust⁹ (black carbon or “soot,” a component of light absorbing carbon). These types of sources are often located within more populated areas, thus leading to increased public exposure to their health damaging emissions.

Examples of measures that can affect organic and light absorbing carbon PM_{2.5} levels include reducing emissions from residential and commercial wood combustion, reducing diesel exhaust emissions from mobile and stationary sources, and addressing volatile organic compound (VOC) emissions from sources such as gasoline storage tanks. Measures that can affect nitrate PM_{2.5} include further reducing oxides of nitrogen (NO_x) from electric power plants and light-duty vehicles. Co-benefit opportunities also exist. For example, measures to reduce ground-level ozone can also lead to decreases in PM_{2.5}

⁵ USEPA (U.S. Environmental Protection Agency). (2009). *Integrated Science Assessment for Particulate Matter*, EPA/600/R-08/139F, December 2009.

⁶ USEPA (U.S. Environmental Protection Agency). (2012). *Particulate Matter (PM) Regulatory Actions*, <http://www.epa.gov/pm/actions.html>.

⁷ NESCAUM. (2012). *Contribution of Non-Sulfate Aerosols to MANE-VU Regional Haze*, Technical Memorandum to MANE-VU, January 2012. Available at <http://www.nescaum.org/documents/nonso4-haze-manevu-final-20120131.pdf>.

⁸ See, e.g., Allen, G.A., P.J. Miller, L.J. Rector, M. Brauer, and J.G. Su. (2011). “Characterization of Valley Winter Woodsmoke Concentrations in Northern NY Using Highly Time-Resolved Measurements.” *Aerosol and Air Quality Research*, **11**, 519–530, doi:10.4209/aaqr.2011.03.0031.

⁹ See, e.g., NESCAUM. (2012). *Air Quality, Electricity, and Back-up Stationary Diesel Engines in the Northeast*, NESCAUM, Boston, MA, August 2012. Available at http://www.nescaum.org/documents/nescaum-aq-electricity-stat-diesel-engines-in-northeast_20120801.pdf.

because of the roles ozone and its precursor NO_x have in promoting the formation of organic carbon and nitrate PM_{2.5}.

The next section of this report provides a status update of progress achieved to date towards the national visibility goal and PM_{2.5} health standards, and an overview of the largest emission sectors in the MANE-VU region contributing to PM_{2.5} pollution. This is followed by a review of recent scientific studies to help provide additional context for non-sulfate control strategies. Finally, a summary of several non-sulfate control strategies is presented. Identifying other options, and quantifying the extent of all potential reductions and their impacts on visibility and public health in the MANE-VU region will be an important part of future work in developing “beyond sulfate” strategies to achieve air quality goals.

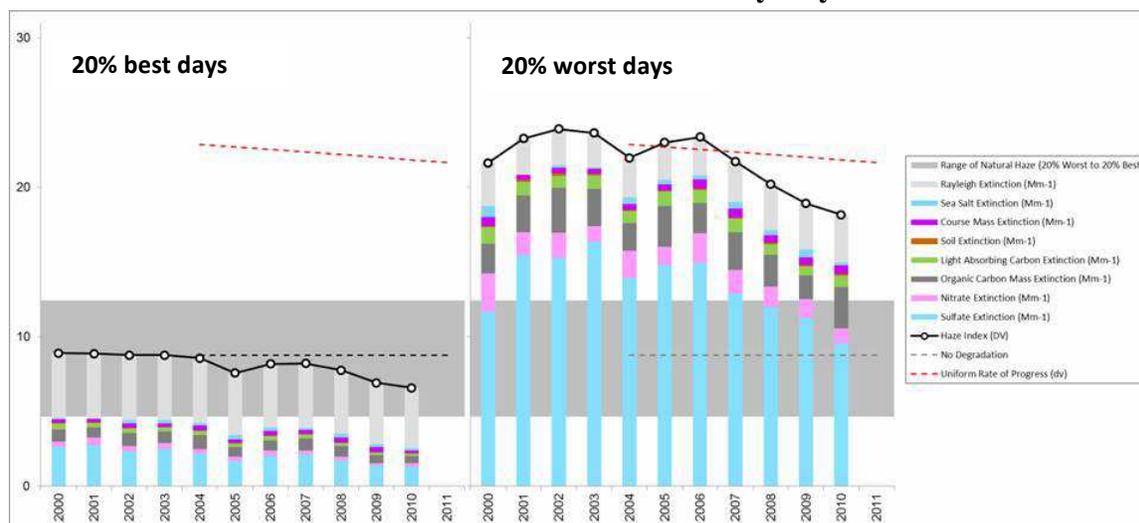
2. OVERVIEW OF VISIBILITY AND PM_{2.5} MASS IN MANE-VU REGION

2.1. Visibility Trends

Long-term monitoring data collected by the Interagency Monitoring of Protected Visual Environments (IMPROVE) network indicate a general improving trend in visibility at MANE-VU Class I areas over the past two decades. Decreasing trends were seen in sulfate, nitrate, and total carbon concentrations across all MANE-VU sites when analyzing IMPROVE data over periods as long as twenty years (1989-2008).¹⁰ This suggests that local and regional multipollutant measures in the eastern United States are leading to downward trends across most components of visibility-impairing aerosols at the MANE-VU Class I sites.

As an example of visibility improvements since the promulgation of the Regional Haze Rule in 1999, Figure 2-1 shows visibility trends (in units of inverse megameters – Mm⁻¹) at Acadia National Park, ME on the 20 percent best (left side) and worst (right side) visibility days. The trends, which are generally similar at all MANE-VU Class I areas, show a continuing large contribution to visibility impairment from sulfate, while its decrease over time has led to visibility improvements on both the best and worst days. With decreasing sulfate levels, other pollutant components are becoming relatively greater contributors to visibility impairment, notably nitrate and carbon (light absorbing and organic) aerosols. Note that the Rayleigh and sea salt components are largely a part of natural background visibility (i.e., not a target of pollution control strategies).

Figure 2-1. Contributions to Regional Haze for Acadia National Park, ME on 20% Best and Worst Visibility Days



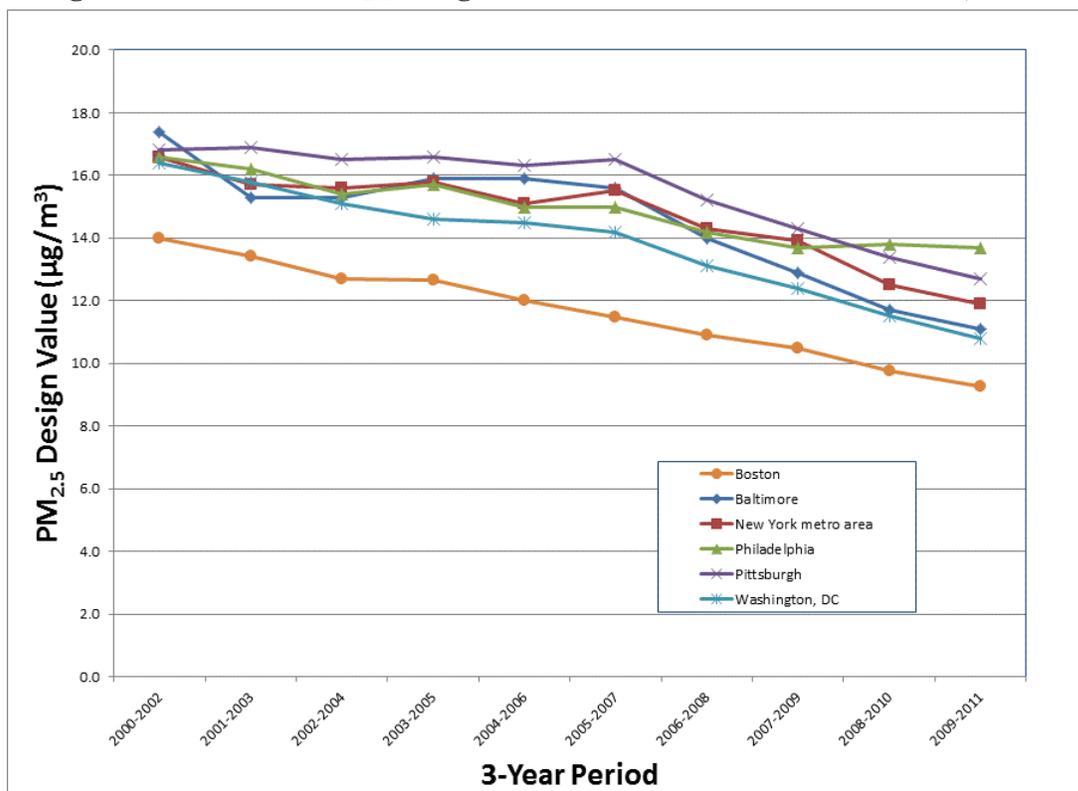
¹⁰ IMPROVE (Interagency Monitoring for Protected Visual Environments) Report V. (2011). *Spatial and Seasonal Patterns and Temporal Variability of Haze and its Constituents in the United States*. Prepared by Hand, J.L. et al. ISSN 0737-5352-87 (June). Available at <http://vista.cira.colostate.edu/improve/publications/Reports/2011/2011.htm>.

Similar decreasing long-term trends were also seen in a multi-year (1988-2005) study of modeled sulfate and nitrate deposition compared with observations in the northeastern United States. These trends are attributable to a number of sulfur dioxide and NO_x reduction programs during the study period, including the Clean Air Act Acid Rain Program, the Ozone Transport Commission (OTC) NO_x Budget Program, the NO_x SIP Call, and mobile source measures.¹¹

2.2. PM_{2.5} Mass and NAAQS Trends

As with the visibility trends in MANE-VU’s Class I areas, PM_{2.5} mass as reflected in air quality trends in MANE-VU urban areas has also been declining since 2000. Figure 2-2 shows the declining trends in annual primary NAAQS PM_{2.5} design values (3-year annual averages) for several metropolitan areas in the MANE-VU region: Baltimore, MD; Boston, MA; New York City metro area, NY/NJ; Philadelphia, PA; Pittsburgh, PA; and Washington, DC.¹²

Figure 2-2. Annual PM_{2.5} Design Value Trends in MANE-VU Cities, 2000-2011



¹¹ Civerolo, K., C. Hogrefe, E. Zalewsky, W. Hao, G. Sistla, B. Lynn, C. Rosenzweig, and P.L. Kinney. (2010). “Evaluation of an 18-year CMAQ simulation: Seasonal variations and long-term temporal changes in sulfate and nitrate.” *Atmos. Environ.*, **44**, 3745-3752, doi: 10.1016/j.atmosenv.2010.06.056.

¹² Design values from USEPA, Air Trends, Design Values, at <http://www.epa.gov/airtrends/values.html>.

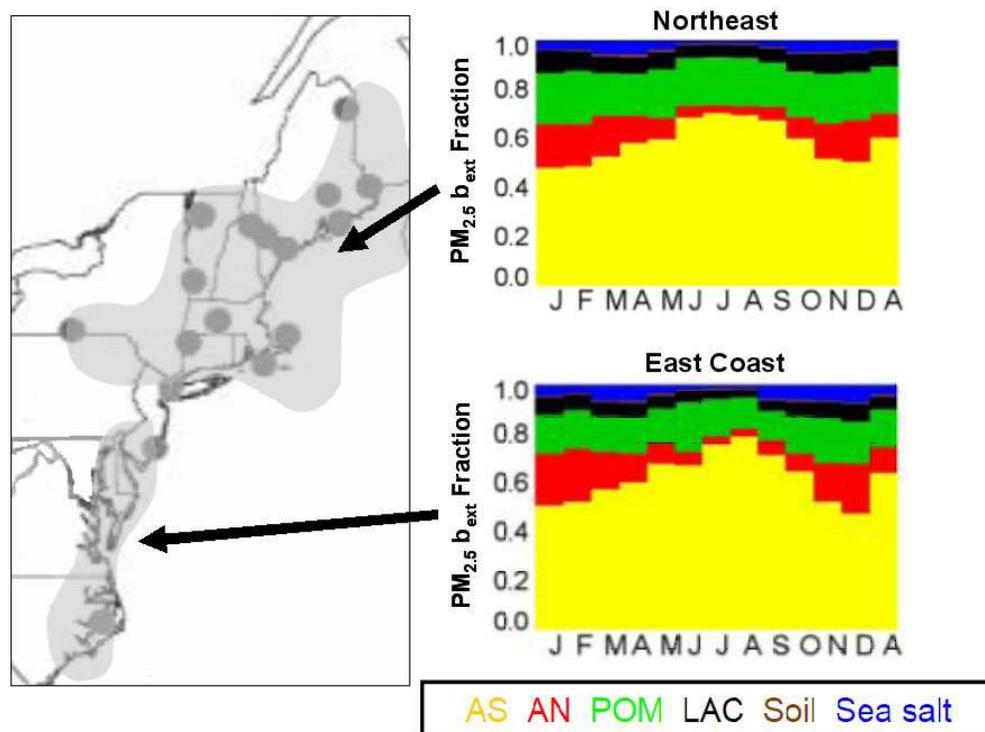
2.3. Component Contributions to Light Extinction and Total PM_{2.5} Mass in MANE-VU Region

2.3.1. Light extinction in MANE-VU Class I areas

Figure 2-3 shows monthly and annual contributions of PM_{2.5} components to light extinction in two areas within the MANE-VU region.¹⁰ The “East Coast” region of the figure’s map includes the Brigantine, NJ Class I area, and the “Northeast” region includes all the other MANE-VU Class I areas. The two regions show similar monthly and annual contributions from the different PM_{2.5} components.

As seen previously in the visibility trends of Figure 2-1, the largest contributor to visibility impairment is sulfate, followed by the carbon components and nitrates. The nitrate contribution is relatively greater in the colder months, reflecting the greater chemical bond stability of ammonium nitrate at lower temperatures.

Figure 2-3. Rural IMPROVE sites 2005-2008 regional monthly mean PM_{2.5} light extinction coefficient (b_{ext}) fractions for MANE-VU region



Note: Figure 2-3 is adapted from Figure 5.1.12 in IMPROVE Report V (2011).¹³ The colored bars show regional monthly mean (2005–2008) PM_{2.5} light extinction coefficient (b_{ext}) fractions for the two shaded regions of the map, based on IMPROVE rural monitors (shown as dots). The letters on the x-axis correspond to the month and “A” corresponds to “annual” mean. Ammonium sulfate (AS) is in yellow,

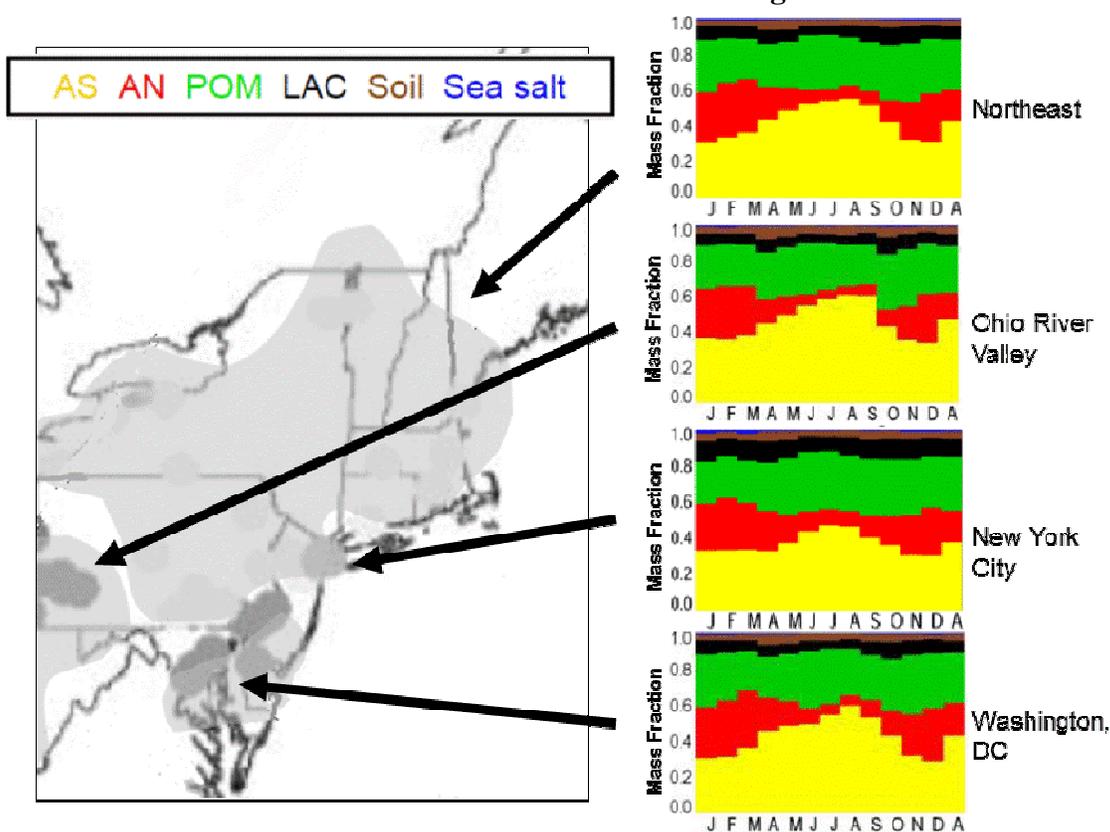
¹³ IMPROVE (Interagency Monitoring for Protected Visual Environments) Report V. (2011). *Spatial and Seasonal Patterns and Temporal Variability of Haze and its Constituents in the United States*. Prepared by Hand, J.L. et al. ISSN 0737-5352-87 (June). Available at <http://vista.cira.colostate.edu/improve/publications/Reports/2011/2011.htm>.

ammonium nitrate (AN) in red, particulate organic matter (POM) in green, light absorbing carbon (LAC) in black, soil in brown, and sea salt in blue.

2.3.2. Fine particle mass fractions in MANE-VU urban areas

The species contributions to total $PM_{2.5}$ mass are shown in Figure 2-4, based on monitoring data from the Chemical Speciation Network (CSN). As with visibility contributions, sulfate is the largest contributor to total $PM_{2.5}$ mass in urban areas of the MANE-VU region. Sulfate's relative mass contribution, however, is less than its contribution to light extinction in rural areas of MANE-VU. For mass contributions, the nitrate (particularly in winter) and carbon components contribute a relatively larger share to the monthly mean mass concentrations compared to their light extinction contributions.

Figure 2-4. Urban CSN sites 2005-2008 regional monthly mean $PM_{2.5}$ reconstructed mass fractions for MANE-VU region



Note: Figure 2-4 is adapted from Figure 4.1.16 in IMPROVE Report V (2011).¹⁴ The colored bars show regional monthly mean (2005–2008) reconstructed $PM_{2.5}$ mass fractions for the shaded regions of the map, based on Chemical Speciation Network (CSN) urban monitors (shown as dots). The letters on the x-axis correspond to the month and “A” corresponds to “annual” mean. Ammonium sulfate (AS) is in yellow, ammonium nitrate (AN) in red, particulate organic matter (POM) in green, light absorbing carbon (LAC) in black, soil in brown, and sea salt in blue.

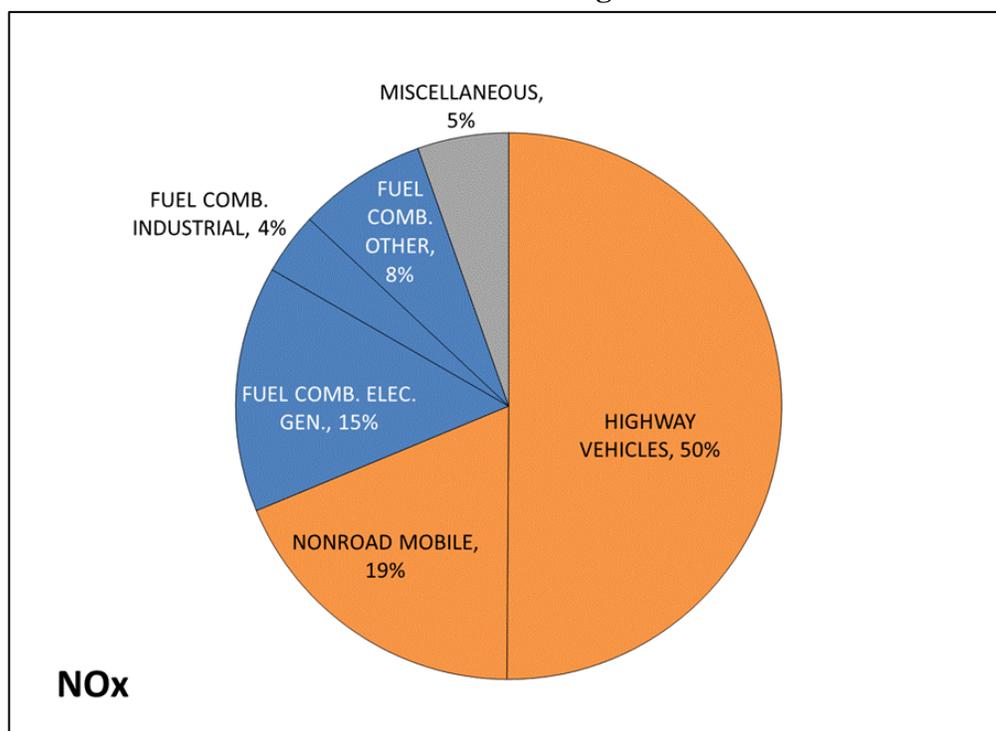
¹⁴ IMPROVE (Interagency Monitoring for Protected Visual Environments) Report V. (2011). *Spatial and Seasonal Patterns and Temporal Variability of Haze and its Constituents in the United States*. Prepared by Hand, J.L. et al. ISSN 0737-5352-87 (June). Available at <http://vista.cira.colostate.edu/improve/publications/Reports/2011/2011.htm>.

2.4. Overview of MANE-VU Emissions

As seen in the light extinction and fine mass contributions described in Section 2.3, the largest contributors to visibility impairment and PM_{2.5} mass after sulfates in the MANE-VU region arise from NO_x, VOCs, and primary PM_{2.5} emissions. Emissions of NO_x contribute to nitrates, and also can help promote the formation of secondary organic aerosols through atmospheric chemistry. Emissions of VOCs can also lead to formation of secondary organic aerosols. Primary PM_{2.5} contributes to light absorbing carbon (e.g., diesel soot) or organic aerosols (e.g., wood smoke).

The following figures show the source sector contributions within the MANE-VU region for emissions of NO_x, VOCs, and primary PM_{2.5} for the year 2007.¹⁵ The figures provide a general overview of the types of emission source sectors for the different air pollutants, and their relative contributions to the total emissions. This in turn suggests where to target non-sulfate pollution control strategies in order to have the biggest relative impacts on visibility impairment and PM_{2.5} mass in the MANE-VU region.

Figure 2-5. 2007 NO_x Emission Source Contributions in MANE-VU Region



In the 2007 MANE-VU inventory, total NO_x emissions for the MANE-VU region were about 2.3 million tons. As seen in Figure 2-5, highway vehicles are the largest source of NO_x emissions in the MANE-VU region, followed by nonroad vehicles and fossil fuel combustion for electricity generation.

¹⁵ Mid-Atlantic Regional Air Management Association. (2012). *2007 Regional Emissions Inventory Version 3*, available at <http://marama.org/technical-center/emissions-inventory/2007-emissions-and-projections/version-3-2007-emissions-inventory> (accessed November 28, 2012).

Figure 2-6. 2007 VOC Emission Source Contributions in MANE-VU Region, Including Biogenic VOCs

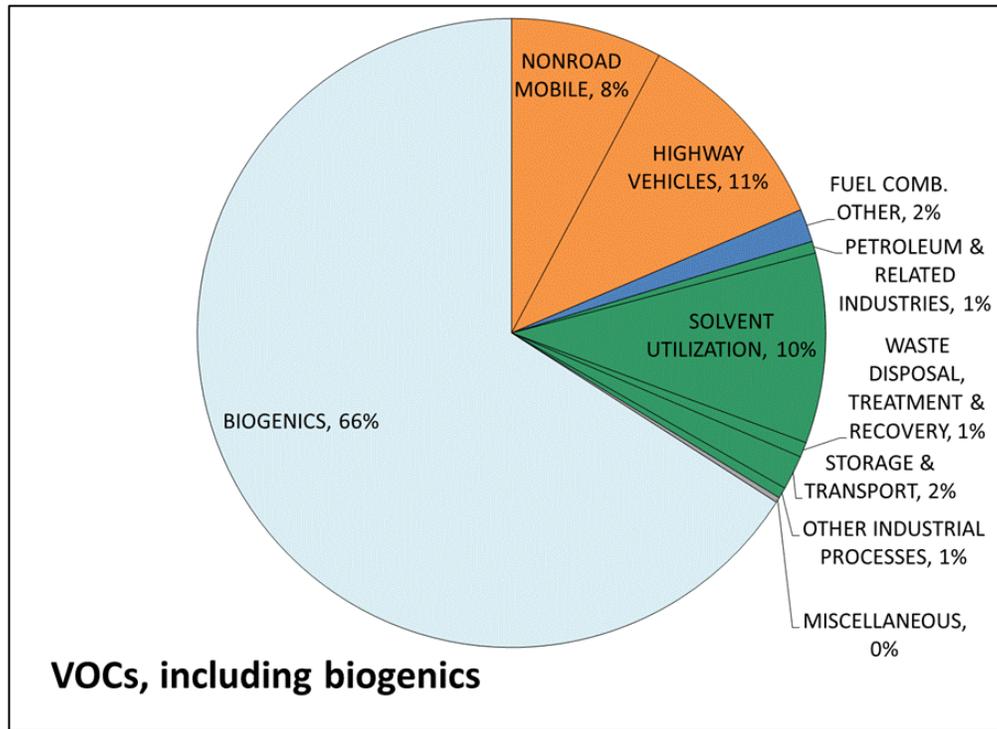
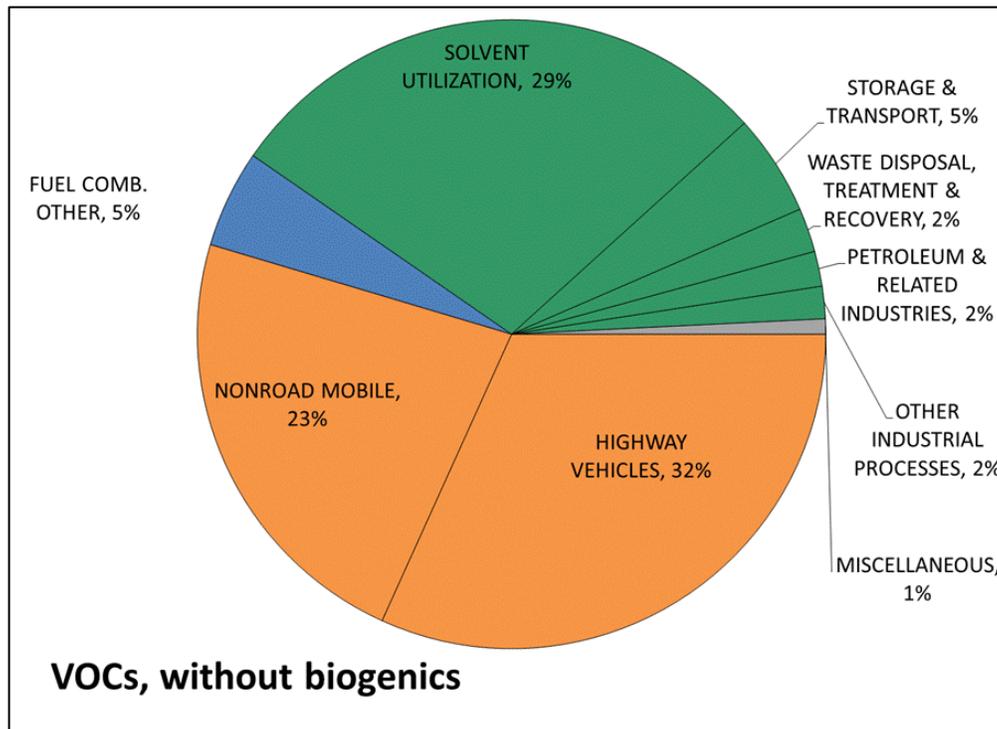
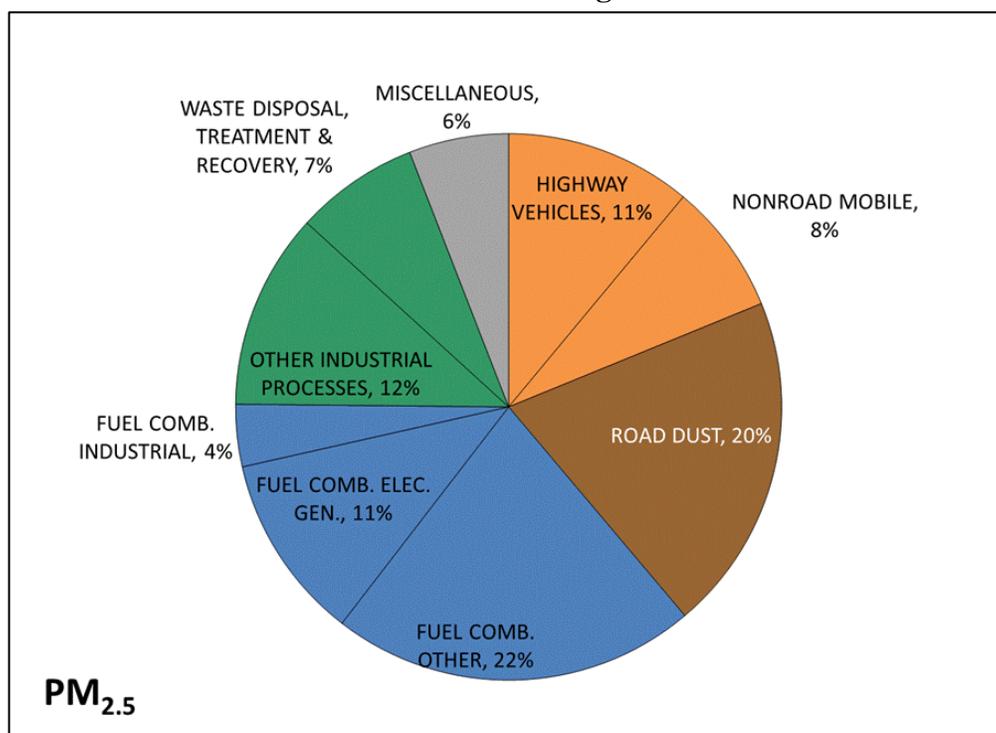


Figure 2-7. 2007 VOC Emission Source Contributions in MANE-VU Region, Excluding Biogenic VOCs



In the 2007 inventory, total VOC emissions, including biogenic, in the MANE-VU region amounted to over 5.5 million tons. For total VOC emissions, biogenic emissions (e.g., from vegetation) are the largest source of VOCs in the MANE-VU region, comprising about two-thirds of the total VOC inventory (Figure 2-6). Excluding biogenic VOCs, the anthropogenic portion of the 2007 MANE-VU VOC inventory is about 1.9 million tons. The largest source of non-biogenic VOC emissions in the region is from highway vehicles, followed by solvent use and nonroad mobile sources (Figure 2-7). As discussed later in the literature review of Section 3, while secondary carbon aerosols may have a large biogenic VOC source component, this significant fraction of $PM_{2.5}$ mass is not entirely “natural,” and may be controllable through strategies aimed at other air pollutants that promote secondary organic aerosol formation.

Figure 2-8. 2007 Primary $PM_{2.5}$ Emission Source Contributions in MANE-VU Region



In 2007, primary $PM_{2.5}$ emissions totaled about 485,000 tons in the MANE-VU region. The largest source sector of primary $PM_{2.5}$ is from fuel combustion other than for electricity generation (Figure 2-8), a category that encompasses commercial, institutional, and residential combustion sources (e.g., wood combustion for space heating). Road dust is the second largest source of $PM_{2.5}$, followed by other industrial processes and fossil fuel combustion for electricity generation. The collective sectors covering nonroad mobile sources and highway vehicles (e.g., mobile source diesel engines) are about equal to estimated road dust emissions, the second largest category.

An analysis of IMPROVE data indicates that concentrations of primary $PM_{2.5}$ (e.g., light absorbing carbon) have been decreasing in the eastern United States due to

introduction of cleaner engines, residential wood burning technologies, and prescribed burning practices.¹⁶

2.5. Contributions Based on Source Receptor Methods

Previous work by MANE-VU summarized the results of receptor model results for rural and urban sites in and outside the MANE-VU region to identify common source categories that had discernible impacts on PM_{2.5} mass concentrations and visibility impairment.¹⁷ Receptor-based models use ambient measurement data at one or more receptor locations and work “backward” to identify sources contributing to historical ambient pollutant concentrations at the receptor locations.

The summarized results of the receptor modeling studies are consistent with the aerosol component contributions and emissions inventory information presented in the preceding subsections. The studies collectively identified sulfate aerosols from coal burning as the largest mass-contributing and visibility-impairing source category at most sites. Nitrate aerosols were small to moderate contributors to average fine mass, with significant local source contributions in urban areas. Mobile sources and secondary organic aerosols were moderate to large contributors to average fine mass, with gasoline and diesel vehicles important in urban areas, and biogenic organics likely more important contributors in rural areas. Wood smoke was a small to moderate contributor to fine mass, with higher contributions typically found in rural areas compared to urban, and higher winter peaks in northern areas from residential wood combustion.

¹⁶ Chen, L.-W.A., J.C. Chow, J.G. Watson, and B.A. Schichtel. (2012). “Consistency of long-term elemental carbon trends from thermal and optical measurements in the IMPROVE network.” *Atmos. Meas. Tech.*, **5**, 2329-2338, doi: 10.5194/amt-5-2329-2012.

¹⁷ MANE-VU. (2006). *Contributions to Regional Haze in the Northeast and Mid-Atlantic United States*. Prepared by NESCAUM, Boston, MA (August), see Appendix B. Available at <http://www.nescaum.org/documents/contributions-to-regional-haze-in-the-northeast-and-mid-atlantic--united-states/>.

3. INFLUENCE OF ANTHROPOGENIC EMISSIONS ON BIOGENIC ORGANIC AEROSOLS

During the 65 years in which the USEPA and regional planning organizations aim to reach the national visibility goal, opportunities will arise during state planning processes to revise the basic control approach.¹⁸ This enables new scientific results to continue to exert a positive influence as states implement new regulatory control programs for multiple pollutants, and as ambient concentrations of these pollutants change relative to each other over time. As these relationships between species change, changes in atmospheric chemistry may dictate revised control approaches.

A currently evolving area is the interplay of anthropogenic emissions and biogenic secondary organic aerosol (SOA) formation. Plant matter contributes more than two thirds of global VOC emissions to the atmosphere. Therefore SOA is predominantly derived from biogenic VOCs. This is supported by radiocarbon dating of ambient organic aerosol, which shows that SOA consists primarily of non-fossil, modern carbon, particularly in summer. This same SOA, however, correlates strongly with indicators of anthropogenic pollution, such as CO and alkyl nitrates.¹⁹

Modeling and experimental studies indicate that anthropogenic NO_x and primary carbonaceous emissions can affect formation of biogenic SOAs through various mechanisms.²⁰ A literature review by Hoyle *et al.* (2011) concluded the published data supported the existence of an enhancement effect by anthropogenic emissions upon the formation of biogenic SOAs.²¹ These collective studies challenge the implicit assumption that, because its components are naturally emitted, biogenic SOAs are not controllable through conventional air quality management.

In modeling the anthropogenic influence on biogenic SOA, Carlton *et al.* (2010) estimated that more than 50 percent of biogenic SOA in the eastern U.S. could be controlled.¹⁹ Modeled biogenic SOA was most sensitive to controllable primary carbonaceous particulate matter and NO_x. Reductions in NO_x decreased oxidant levels and, consequently, the formation of semivolatile species from VOC oxidation.

¹⁸ Watson, J. (2002). "Visibility: Science and Regulation," *J. Air & Waste Manage. Assoc.*, **52**, 628-713.

¹⁹ Carlton, A.G., R.W. Pinder, P.V. Bhave, and G.A. Pouliot. (2010). "To What Extent Can Biogenic SOA be Controlled?" *Environ. Sci. Technol.*, **44**, 3376-3390, doi: 10.1021/ES903506B.

²⁰ See, e.g., Zaveri, R.A. *et al.* (2010). "Nighttime chemical evolution of aerosol and trace gases in a power plant plume: Implications for secondary organic nitrate and organosulfate aerosol formation, NO₃ radical chemistry, and N₂O₅ heterogeneous hydrolysis." *J. Geophys. Res.*, **115**, D12304, doi: 10.1029/2009JD013250; Carlton, A.G., R.W. Pinder, P.V. Bhave, and G.A. Pouliot. (2010). "To What Extent Can Biogenic SOA be Controlled?" *Environ. Sci. Technol.*, **44**, 3376-3390, doi: 10.1021/ES903506B; Hogrefe, C., S.S. Isukapalli, X. Tang, P.G. Georgopoulos, S. He, E.E. Zalewsky, W. Hao, J.-Y. Ku, T. Key, and G. Sistla. (2011). "Impact of Biogenic Emission Uncertainties on the Simulated Response of Ozone and Fine Particulate Matter to Anthropogenic Emission Reductions." *J. Air & Waste Manage. Assoc.*, **61**, 92-108, doi: 10.3155/1047-3289.61.1.92; Perraud, V. *et al.* (2011). "Nonequilibrium atmospheric secondary organic aerosol formation and growth." *PNAS*, **109**, 2836-2841, doi: 10.1073/pnas.1119909109.

²¹ Hoyle, C.R., M. Boy, N.M. Donahue, J.L. Fry, M. Glasius, A.G. Hallar, K.H. Hartz, M.D. Petters, T. Petäjä, T. Rosenoern, and A.P. Sullivan. (2011). "A review of the anthropogenic influence on biogenic secondary organic aerosol." *Atmos. Chem. Phys.*, **11**, 321-343, doi: 10.5194/ACP-11-321-2011.

Reductions in primary carbonaceous particulate matter had the largest effect by limiting the organic material available for condensable semivolatile species.

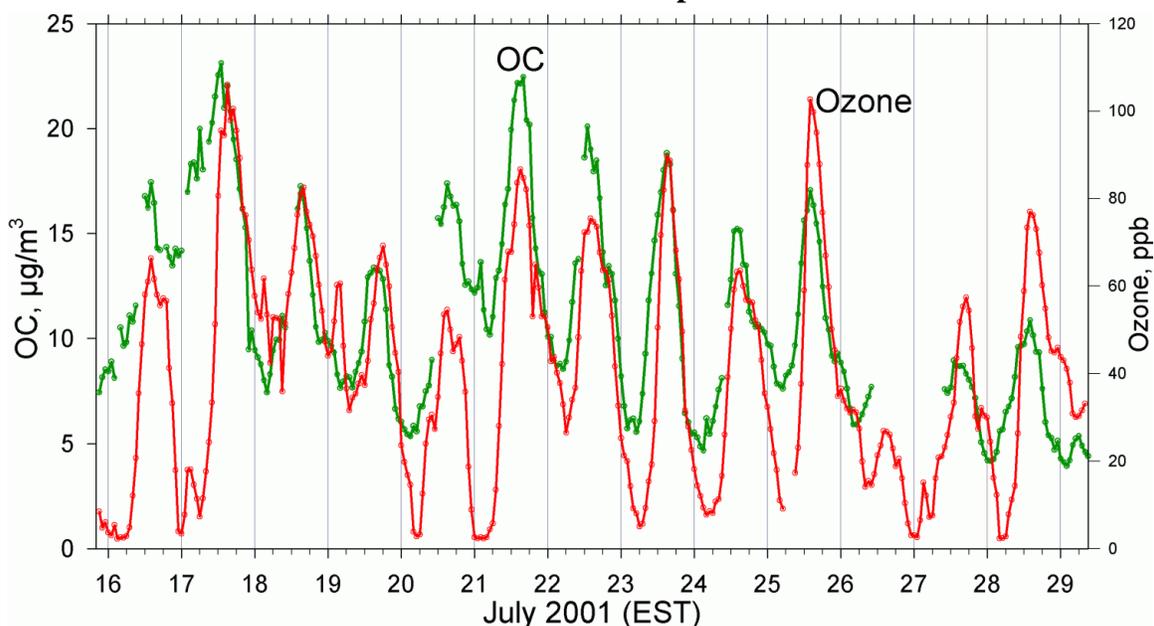
Tsimpidi *et al.* (2008) estimated the impact of anthropogenic VOC and NO_x emissions on particulate matter and its components in the eastern U.S. using a three-dimensional chemical transport model.²² The study simulated air quality management resulting in 50 percent reductions in NO_x and 50 percent reductions of anthropogenic VOC emissions. It was found that the reduction of NO_x emissions affects the oxidant levels and consequently affects both the anthropogenic and biogenic SOA formation. VOC reductions were found to decrease SOA formation in the summer, but increase concentrations of other components of fine particulate matter.

Hand *et al.* (2011) noted similar annual fluctuations in the impacts of anthropogenic emissions on SOA, stressing the importance of seasonality in determining the sources of SOA formation.²³ This study combined monitoring data from the IMPROVE network and the USEPA's urban/suburban CSN network. Urban and rural particulate concentrations were highest during the same season in some regions of the U.S., including the East, and during opposite seasons in other regions, in particular the Southwest. This was attributed to biomass smoke and biogenic emissions in summer months, particularly in rural areas in the West. Primary sources and regional transport were main contributors in the East.

²² Tsimpidi, A.P., V.A. Karydis, and S.N. Pandis. (2008). "Response of Fine Particulate Matter to Emission Changes of Oxides of Nitrogen and Anthropogenic Volatile Organic Compounds in the Eastern United States." *J. Air & Waste Manage. Assoc.*, **58**, 1463-1473, doi: 10.3155/1047-3289.58.11.1463.

²³ Hand, J.L., B.A. Schichtel, M. Pitchford, W.C. Malm, and N.H. Frank. (2012). "Seasonal composition of remote and urban fine particulate matter in the United States." *J. Geophys. Res.*, **117**, D05209, doi: 10.1029/2011JD017122.

Figure 3-1. Hourly Organic Carbon (OC) Aerosol and Ozone, Summer 2001, Northeast Philadelphia



Monitoring data collected within the MANE-VU region also suggest a correlation between oxidant levels (e.g., ozone) and SOA formation. Figure 3-1 shows a two-week time-series plot of hourly ozone and organic carbon (OC) aerosol concentrations from a monitoring site nine miles northeast of downtown Philadelphia.²⁴ Figure 3-1 shows OC aerosol closely tracking ozone concentrations, including the rapid drop in late afternoons. The removal of ozone by deposition and titration in the early evening is well understood, but OC does not have these rapid removal mechanisms. This implies that a large portion of the observed mid-day OC peak is fresh (local, urban) SOA, and the rapid afternoon drop in OC is due to a phase change of the fresh SOA back to gas phase organics, not removal of organic carbon from the atmosphere. In other words, ozone and related oxidants push gas-phase VOC into the aerosol phase (SOA); when these oxidants are removed, the equilibration shifts from SOA back to gas phase VOC. This tracking of OC with ozone would be strongest with fresh SOA; photochemically aged (rural) SOA is more stable and would not be expected to show this pattern.

²⁴ NESCAUM. (2010). *Semi-continuous Aerosol Carbon Data from the MANE-VU RAIN Program: Assessment of Data Quality and Data Analysis Approaches*, prepared for MANE-VU (February). Available at <http://www.nescaum.org/documents/manevu-sunset-carbon-report-feb2010-final.pdf/>.

4. NON-SULFATE STRATEGIES

Section 2 reviewed the relative contributions of non-sulfate particulate matter concentrations in the MANE-VU region and the emission source sectors contributing to their presence in the atmosphere. Section 3 gave an overview of the scientific literature on the interplay of NO_x and carbonaceous particulate matter in contributing to secondary organic aerosols. This information provides the context for considering potential future strategies to address the non-sulfate portion of particulate matter affecting visibility and public health in the MANE-VU region.

In general, the non-sulfate particulate constituents having the greatest impact in the MANE-VU region after sulfates are nitrates and carbonaceous particulates (organic and light absorbing carbon). The literature review suggests that reductions of NO_x to reduce the nitrate component will also help reduce the carbonaceous portion. This is especially salient in the context of secondary organic aerosols that appear to have a major biogenic carbon component, but whose formation is fostered by atmospheric chemistry involving NO_x.

The following subsections give overviews of non-sulfate control options for NO_x and VOC emission sources contributing to non-sulfate particulate matter in the region. While the options are generally separated into separate NO_x and carbon aerosol control strategies, it should be recognized that many measures reducing one type of pollutant can also reduce other pollutants as well (although the opposite can also occur). Non-traditional “control” options such as energy efficiency and combined heat and power can also reduce multiple pollutants through reduced energy consumption.

4.1. NO_x reduction strategies

4.1.1. Mobile sources

NO_x emissions contribute to PM_{2.5} loadings through formation of nitrate aerosols and in promoting the formation of SOA. The largest source of NO_x emissions within the MANE-VU region is highway vehicles, which comprise over 40 percent of the total 2008 regional NO_x inventory (*see* Figure 2-5). Under the federal Clean Air Act, the MANE-VU states have two options for controlling passenger vehicle NO_x emissions. These are either the federal highway vehicle emission standards promulgated by the USEPA, or opting into the California low emission vehicle (LEV) program, which historically has been more stringent than the federal standards.²⁵

California low emission vehicle program

All states in the MANE-VU region with the exception of New Hampshire have adopted the California LEV program in lieu of the federal vehicle standards. Since the inception of its first low emission vehicle program (“LEV I”), California has over time revisited the program twice and increased the stringency of its requirements in recognition of the need to continue progress towards meeting public health air quality

²⁵ Miller, P. and M. Solomon. (2009). “A Brief History of Technology-Forcing Motor Vehicle Regulations.” *EM*, pp. 4-8 (June 2009).

standards. When California revises its LEV program, the states that have opted into it must also revise their own state rules to maintain consistency with California standards. The most recent revision to California's program is LEV III, which the state adopted in early 2012. As part of California's waiver request to the USEPA, the state projects that LEV III will achieve an approximate 75 percent reduction in combined NO_x and non-methane organic gas (NMOG) fleet average emissions from new light duty vehicles by 2025 compared to 2010.²⁶

Federal Tier 3 program

The USEPA has indicated a need to revise federal passenger vehicle standards in order to help areas of the country meet increasingly stringent ambient air quality standards.²⁷ This revision, called "Tier 3," would also lower gasoline sulfur content in addition to the change in new vehicle emission standards. While most MANE-VU states have adopted the California LEV program, the implementation of a new federal Tier 3 program can lead to additional NO_x reductions within and outside of the MANE-VU region from the existing motor vehicle fleet. Sulfur impairs ("poisons") the catalyst performance of motor vehicle tailpipe controls, so lowering sulfur content in gasoline lowers NO_x emissions from the existing vehicle fleet by improving catalyst performance.

If gasoline fuel sulfur content was lowered to 10 parts per million (ppm), it would reduce NO_x emissions by approximately 25 percent from much of the existing national fleet of gasoline-powered vehicles.²⁸ The estimated per state and aggregate regional reduction benefits for gasoline-powered onroad vehicles in the eastern U.S. are presented in Table 4-1.²⁹ Based on the preliminary regional mobile source inventory for 2017, as calculated by NESCAUM with the USEPA's MOVES model, a 10 ppm average gasoline sulfur requirement would reduce NO_x emissions in the MANE-VU region by almost 50,000 tons per year, or 136 tons per day.

As shown in Table 4-1, a 10 ppm low sulfur gasoline component in the Tier 3 program would reduce NO_x emissions by more than 60,000 tons per year in eight Midwest states and almost 67,000 tons per year in 10 southeastern states that abut the MANE-VU region. These reductions will benefit air quality in the MANE-VU region by: (1) lowering the "ozone reservoir" that forms in the eastern U.S., (2) reducing the amount of low-level NO_x emissions and pollutants derived from NO_x (e.g., nitrates) that are transported into the MANE-VU region, (3) reducing oxidant levels that promote SOA

²⁶ California Air Resources Board. (2012). *Clean Air Act § 209(b) Waiver Support Document Submitted by the California Air Resources Board*, submitted to USEPA, May 2012 (EPA Docket ID: EPA-HQ-OAR-2012-0562-0004).

²⁷ Federal Register. (2011). *Unified Agenda 2060-AQ86: Control of Air Pollution From Motor Vehicles: Tier 3 Motor Vehicle Emission and Fuel Standards*, Unified Agenda current as of Fall 2011, available at <https://www.federalregister.gov/regulations/2060-AQ86/control-of-air-pollution-from-motor-vehicles-tier-3-motor-vehicle-emission-and-fuel-standards> (accessed September 12, 2012).

²⁸ Based on Ball, D., D. Clark, and D. Moser. (2011). *Effects of Fuel Sulfur on FTP NO_x Emissions from a PZEV 4 Cylinder Application*, SAE Technical Paper 2011-01-0300, doi: 10.4271/2011-01-0300.

²⁹ NESCAUM. (2011). *Assessment of Clean Gasoline in the Northeast and Mid-Atlantic States*, NESCAUM, Boston, MA (November). Available at <http://www.nescaum.org/documents/nescaum-tier-3-low-s-gasoline-20111121.pdf>.

formation, and (4) ensuring that vehicles registered in other states but operating in the MANE-VU states emit less NO_x.

Light-duty vehicles in the three regions are predicted to emit over 1,100,000 tons of NO_x in 2017 absent a 10 ppm low sulfur gasoline standard. A 10 ppm average sulfur gasoline requirement could reduce NO_x emission from this sector by nearly 180,000 tons per year, or almost 500 tons per day in the eastern U.S.

Table 4-1. Onroad Gasoline Vehicle Emissions and Estimated Reductions

State/DC/Region	2017 Gasoline Onroad Base NO _x (tpy)	Est. NO _x Reductions from 10 ppm S	
		(tpy)	(tpd)
Connecticut	20,700	-3,100	-8
Delaware	5,400	-800	-2
District of Columbia	2,000	-300	-1
Maine	10,000	-1,500	-4
Maryland	32,600	-5,000	-14
Massachusetts	35,100	-5,300	-15
New Hampshire	8,400	-1,300	-4
New Jersey	44,300	-6,700	-18
New York	88,600	-13,500	-37
Pennsylvania	70,500	-10,700	-29
Rhode Island	5,600	-900	-2
Vermont	5,000	-800	-2
MANE-VU Total	328,200	-49,900	-136
Midwest States Total³⁰	402,300	-61,000	-167
Southeast States Total³¹	439,100	-66,600	-183
3 Region Total	1,169,600	-177,500	-486

4.1.2. MANE-VU stationary source NO_x control options

The MANE-VU states have recently considered at least four potential measures to further reduce NO_x from stationary sources in the region. These would reduce emissions from:

- Stationary Generators;
- Natural Gas-Fired Industrial, Commercial, and Institutional Boilers, Steam Generators, Process Heaters, and Water Heaters;
- High Electric Demand Day Combustion Turbines (HEDDCT);
- Oil and Gas Boilers Serving EGUs.

The potential NO_x control approaches are described in an Ozone Transport Commission (OTC) draft technical support document (TSD) summary presented at an

³⁰ IL, IN, IA, MI, MN, MO, OH, WI.

³¹ AL, FL, GA, KY, MS, NC, SC, TN, VA, WV.

OTC meeting in March 2011, and much of the following source summaries are extracted from the draft TSD.³²

Stationary generators

Peaking and baseload non-emergency generators within the region have the potential to emit about 48 tons of NO_x for every hour of operation (based upon the assumption of no controls on the engine and an average emission factor of 32 lb/MWh NO_x). If these peaking and baseload engines were controlled, their emissions could be reduced by approximately 90 percent, which would result in a regional reduction of about 43 tons of NO_x for every hour of operation.

Natural gas-fired industrial, commercial, and institutional boilers, steam generators, process heaters, and water heaters

This would address NO_x emissions from industrial, commercial, and institutional (ICI) boilers, steam generators, process heaters, and water heaters by using ultra low NO_x burners (ULNBs) to control emissions. The estimated achievable NO_x reductions in the region are about 53 tons per day with full implementation of control measures.

High electric demand day combustion turbines (HEDDCT)

For the purpose of this rule, a high electric demand day combustion turbine (HEDDCT) is defined as a 5 to 15 MW or larger (depending on distribution of generating units in individual states) natural gas- or distillate fuel oil-fired combustion turbine that generates and delivers electricity to power the grid for commercial sale, that began operating prior to May 1, 2007 and was operated less than or equal to 50 percent of the time during the ozone seasons of 2007 through 2009. The focus of this rule is on NO_x emissions emitted by HEDDCTs typically for only a few hours a year, but often on the hottest summer days when air quality is poorest. As such, this measure may have little impact on particulate nitrate during cooler periods when electricity demand is less. During the ozone season, it is estimated implementation of this model rule could reduce MANE-VU regional NO_x emissions by 2,500 tons.

Oil and gas boilers serving EGUs

This model rule seeks to regulate oil-fired and gas-fired boilers that provide steam to an electric generating unit with a nameplate capacity of 25 MW or greater, and includes a unit serving a cogeneration facility. The proposed model rule assumes use of low NO_x burners and/or a selective non-catalytic reduction system on existing oil- and gas-fired boilers. These control devices are used widely in industry throughout the United States and are reasonably available given their extensive use. Estimated annual NO_x reductions in the MANE-VU region are about 3,500 tons.

³² Ozone Transport Commission. (2011). *TSD NO_x Section, Draft for Public Comment*, presented at the Ozone Transport Commission (OTC) Stationary Area Source/Modeling/Mobile Source Committee Meeting, Linthicum, MD, March 16, 2011, available at: <http://www.otcair.org/upload/Documents/Meeting%20Materials/TSD%20NOx%20Section%20Public%20Draft%20for%20Comment.pdf>.

4.2. Carbon aerosol reduction strategies

Concentrations of carbon aerosols can be influenced by control of precursor emissions contributing to secondary organic aerosols, or control of primary carbon particulate matter directly emitted by a source. Volatile organic compounds (VOCs), also referred to as hydrocarbons, react in the atmosphere to form secondary organic aerosols via condensation and oxidation processes.

The control of VOCs from pollution sources has long been an active area in state and regional regulatory planning, primarily in strategies to reduce ground level ozone (smog) as well as air toxics (e.g., benzene). Examples include national and state-adopted California motor vehicle tailpipe emission standards, and programs to reduce gasoline evaporation during refueling and from a motor vehicle's fuel system (e.g., Stage I and II controls at gasoline stations; onboard refueling and vapor recovery (ORVR) on cars). Additional efforts within the MANE-VU region have looked at regional coordination and working with the USEPA to reduce VOC emissions from asphalt paving, asphalt production plants, cement kilns, glass furnaces, industrial, commercial, and institutional boilers, small engines (< 50 horsepower), ship lightering, and other sources. These examples of past efforts are not all inclusive, and additional information on measures taken or considered in the MANE-VU region is available on the Ozone Transport Commission website www.otcair.org. In addition, as discussed above, NO_x control strategies can also influence the formation of secondary organic aerosols.

Among primary carbon emission sources, uncontrolled diesel exhaust can be a significant source of black carbon, a component of light absorbing carbon. Monitoring data show that light absorbing carbon has the greatest urban excess over rural for PM_{2.5} components in the eastern United States. Light absorbing carbon also has the sharpest spatial gradients between urban and rural areas, indicating the local influence of urban emissions sources, such as diesel vehicles.³³ Based on these spatial gradients, it can be inferred that non-sulfate strategies aimed at reducing black carbon in diesel exhaust will have the greatest impacts within urban areas where public exposure is also greatest. Diesel exhaust also contains VOCs that can lead to secondary organic aerosols having impacts across a larger regional scale.³⁴

Residential, commercial, and industrial wood combustion are also sources of carbon aerosols, and can be emitted as a complex mix of primary particulate carbon and precursors of secondary organic aerosols. In rural areas, wood combustion can be the major source of carbonaceous particulate matter. In New York State, for example, wood combustion in rural upstate counties is responsible for over 90 percent of the total

³³ IMPROVE (Interagency Monitoring for Protected Visual Environments) Report V. (2011). *Spatial and Seasonal Patterns and Temporal Variability of Haze and its Constituents in the United States*. Prepared by Hand, J.L. et al. ISSN 0737-5352-87 (June). Available at <http://vista.cira.colostate.edu/improve/publications/Reports/2011/2011.htm>.

³⁴ Gentner, D.R. et al. (2012). "Elucidating secondary organic aerosol from diesel and gasoline vehicles through detailed characterization of organic carbon emissions," *PNAS Early Edition*, available at www.pnas.org/cgi/doi/10.1073/pnas.1212272109 (accessed October 25, 2012).

carbonaceous particulate matter.³⁵ Wood smoke emissions can build up to high levels within the more restricted valley topographies found in parts of the MANE-VU region, where population densities can be relatively high.³⁶ Addressing high levels of wood smoke particulate matter within these areas is an important public health concern for a number of states.

4.2.1. MANE-VU VOC control options

The MANE-VU region has recently considered at least four measures for potential state adoption to reduce VOCs. These options would reduce emissions from:

- Stationary Above-Ground Storage Tanks;
- Consumer Products;
- Motor Vehicle and Mobile Equipment Non-assembly Line Coating Operations;
- Architectural, Industrial, and Maintenance Coatings.

The proposed VOC model rules are described in an Ozone Transport Commission (OTC) draft technical support document (TSD) summary presented at an OTC meeting in March 2011.³⁷ The following sections summarizing the proposed model rules are brief extracts taken from the draft TSD. Also included is a description of potential future national tailpipe emission standards for light-duty vehicles coupled with low sulfur gasoline that can further reduce VOCs and other pollutants in the MANE-VU region.

Stationary above-ground storage tanks

The MANE-VU states working through the OTC developed a model rule for public comment in 2011 to address VOCs, such as gasoline, stored in large above-ground stationary storage tanks. These facilities are typically located at refineries, terminals, and pipeline breakout stations. The available control measures are grouped into five categories: deck fittings and seals, domes, roof landings, degassing and cleaning, and inspection and maintenance.

There is some overlap between the model rule and federal standards for storage tanks (e.g., New Source Performance Standards), particularly with regard to deck fittings, seals, and tank inspection requirements, but the federal standards do not generally address roof landings and tank cleaning nor do they require external floating roof tanks to be covered with domes, as the model rule does.

³⁵ New York State Energy Research and Development Authority. (2008). *Assessment of Carbonaceous PM_{2.5} for New York and the Region*. NYSERDA Report 08-01. Prepared by NESCAUM and MJ Bradley & Associates for NYSERDA: Albany, NY (March).

³⁶ New York State Energy Research and Development Authority. (2010). *Spatial Modeling and Monitoring of Residential Woodsmoke across a Non-Urban Upstate New York Region*. Prepared by NESCAUM et al. for NYSERDA: Albany, NY (February).

³⁷ Ozone Transport Commission (OTC). (2011). *TSD VOC Section, Draft for Public Comment*. Presented at the Ozone Transport Commission (OTC) Stationary Area Source/Modeling/Mobile Source Committee Meeting, Linthicum, MD (March 16, 2011). Available at <http://otcair.org/upload/Documents/Meeting%20Materials/TSD%20VOC%20Section%20Public%20Draft%20for%20Comment.pdf> (accessed November 1, 2012).

To reduce VOC emissions from stationary above-ground storage tanks, the OTC model rule proposed the following controls:

- **Deck fittings, seals:** Evaporative VOC losses can occur from deck fittings, particularly slotted guidepoles, and rim seal systems. Control measures include gasketing deck fittings, installing pole sleeves and floats on slotted guidepoles, and gap requirements for rim seals. These measures can result in up to an 80% reduction in standing loss emissions from external floating roof tanks.
- **Domes:** Wind blowing across external floating roof tanks causes evaporative VOC losses. Installing domes on external floating roof tanks can result in about a 60% reduction of remaining VOC emissions after upgrading deck fittings.
- **Roof Landing Controls:** When enough liquid is removed from a floating roof tank such that the roof cannot be lowered farther (i.e., the roof rests on its legs or suspended by cables or hangers), the contact between the floating roof and the VOC liquid is broken as the remaining liquid is removed. This is referred to as a “roof landing.” The vapor space between the floating roof and the liquid surface enables VOC vapors to accumulate and escape from the tank as it sits idle or when refilled. Control options include requiring lander height settings that minimize the vapor space, installation of vapor recovery/control for use when the roof is landed, or modifying the tank to reduce the landed height to one foot or less. The control measures can reduce VOC losses by 60% to 100%, depending on the measure and how the tank is operated.
- **Cleaning and Degassing:** Stationary storage tanks must be cleaned periodically. Before a tank is cleaned, it must be degassed (which is the removal of gases, such as gasoline vapor) so personnel can safely enter to clean the tank and remove accumulated sludge. The sludge removed from the tank can contain residual VOC liquid that may evaporate when exposed to the atmosphere. Measures to reduce VOC evaporation include control of emissions during degassing and controlling exhaust from sludge receiving vessels (such as vacuum trucks). New Jersey has a proposed rule that would require 95% control of emissions during degassing, until the concentration level in a tank is 5,000 parts per million (ppm) as methane, and control of exhaust from the receiving vessel (e.g., vacuum truck).
- **Inspection and Maintenance:** An inspection and maintenance program seeks to reduce VOC emissions by assuring that tank components are in good condition and operating properly. A proposed program in New Jersey for external floating roof tanks would include a full inspection of gap widths for deck fittings and secondary seals annually and of primary seals every five years. Internal floating roof tanks would be inspected annually, with a full inspection of deck fittings and seal gaps each time the tank is emptied and degassed (no less than every 10 years).

Consumer products

The revised OTC model rule for consumer products is based on the California Air Resources Board's (CARB's) 2006 Consumer Products Regulatory Amendments that were adopted by CARB in 2006. The 2006 CARB amendments have more restrictive VOC limits for 13 existing consumer product categories (including subcategories). The

amendments also regulate three new categories (disinfectant, sanitizer, and temporary hair color; including subcategories) for the first time. The revised OTC model rule would achieve VOC reductions through reformulation of the affected product categories by the manufacturers. This may involve switching to a water-based formulation, using an exempt solvent, increasing product solids, or formulating with a non-VOC propellant. Manufacturers can still comply with the proposed model rule through the use of an Innovative Products Exemption (IPE) or an Alternate Control Plan (ACP). The revised OTC model rule for consumer products would apply to anyone who sells, supplies, offers for sale, or manufactures consumer products for use in an OTC member jurisdiction.

Motor vehicle and mobile equipment (MVME) non-assembly line coating operations

The 2009 OTC model rule for Motor Vehicle and Mobile Equipment Non-assembly Line Coating Operations (2009 OTC MVME Model Rule) seeks to limit the VOC content in coatings and cleaning solvents used in motor vehicle and mobile equipment non-assembly line coating operations. Implementation of the model rule would reduce VOC emissions by limiting the VOC content of coatings and cleaning solvents and provide work practice standards for preventing emissions from equipment cleaning and cleaning supply storage.

The 2009 OTC MVME Model Rule applies to people who supply, sell, offer for sale, distribute, manufacture, use or apply automotive coatings and associated cleaning solvents subject to the Model Rule. The model rule limits the VOC content of coatings used in non-assembly line coating operations and limits the VOC content of cleaning solvent to 25 grams per liter. The 2009 OTC MVME Model Rule allows the use of higher VOC content cleaning solutions for special uses and sets lower VOC content limits for many of the formulations resulting from switching from solvent-based formulations to water-based formulations.

Architectural, industrial, and maintenance coatings

The OTC developed its 2002 Architectural and Industrial Maintenance (AIM) Coatings model rule based upon the 2000 CARB Suggested Control Measure (SCM). In 2007, CARB proposed an updated SCM for architectural coatings, which generally lowers VOC emissions through product reformulation and improves definitions of many categories from the 2000 SCM. Of the 47 coating categories regulated in the 2000 SCM, 15 categories have been eliminated (replaced by new categories or deemed unnecessary), 10 categories were added, and 19 have stricter VOC limits. The updated SCM also contains some revised compliance and reporting requirements.

The OTC reviewed the 2007 CARB SCM and found that most of the changes were appropriate for the OTC. The OTC model rule, however, adds some categories that are specific to the OTR, and assigned different limits to three other categories (aluminum roof, bituminous roof, and roof coatings). The OTC model rule is an update of the 2002 Model Rule that has been adopted by most states across the OTR. It includes all the new categories defined in the 2007 CARB SCM as well as the following eight specialty coating categories specific to the OTR:

- Calcimine Recoaters
- Conjugated Oil Varnish (new addition)
- Concrete Surface Retarders
- Conversion Varnish
- Impacted Immersion coatings
- Nuclear Coatings
- Reactive Penetrating Carbonate Stone Sealer (new addition)
- Thermoplastic Rubber Coatings and Mastics

4.2.2. Tier 3 motor vehicle standards and low sulfur gasoline

As previously discussed in the section on NO_x reduction options, the USEPA may propose a “Tier 3” program to strengthen light-duty vehicle emissions standards similar to those of California’s LEV III requirements. Coupled to the change in tailpipe standards would be a lowering of gasoline sulfur content from 30 ppm to 10 ppm. Lowering sulfur content in gasoline immediately improves the efficiency of catalytic converters in the existing motor vehicle fleet by reducing sulfur poisoning of catalyst surfaces. Nationally, the Tier 3 program with low sulfur gasoline could result in a 26 percent decrease in VOC emissions by 2030, along with significant reductions in NO_x and other pollutants.³⁸

4.2.3. Diesel exhaust

While light absorbing carbon in diesel exhaust (i.e., black carbon) appears to have relatively greater impacts in urban areas, recent research also indicates that diesel engine exhaust, particularly from older diesel vehicles, may be a large contributor to secondary organic aerosols as well.³⁹ As a result, diesel exhaust may have a larger regional signature on particulate matter than based on light absorbing carbon alone.

While the USEPA has set more stringent emission limits for newly manufactured engines, a number of project-specific measures have been undertaken in the MANE-VU region to address existing diesel engines that pre-date the more stringent new engine standards. These projects provide funding assistance in a number of areas, such as promoting early retirement of older vehicles or equipment with newer replacements that meet the latest USEPA standards, or retrofitting existing diesel engines with diesel particulate filters or other controls to reduce emissions. Many of the projects have been funded through environmental mitigation settlement agreements arising from government enforcement actions or through annual appropriations under the federal Diesel Emissions Reduction Act (DERA). Many, but not all, of the federally-funded diesel projects have

³⁸ National Association of Clean Air Agencies (NACAA). (2011). *Cleaner Cars, Cleaner Fuel, Cleaner Air: The Need for and Benefits of Tier 3 Vehicle and Fuel Regulations*. NACAA, Washington, DC (October). Available at <http://www.4cleanair.org/Documents/NACAATier3VehandFuelReportFINALOct2011.pdf> (accessed November 1, 2012).

³⁹ Gentner, D.R. *et al.* (2012). “Elucidating secondary organic aerosol from diesel and gasoline vehicles through detailed characterization of organic carbon emissions,” *PNAS Early Edition*, available at www.pnas.org/cgi/doi/10.1073/pnas.1212272109 (accessed October 25, 2012).

been coordinated through two regional collaboratives involving the MANE-VU states, local agencies, and the USEPA. These collaboratives are the Mid-Atlantic Diesel Collaborative⁴⁰ and the Northeast Diesel Collaborative.⁴¹ Going forward, the scale of diesel projects may depend in large part on continued federal funding appropriated under DERA each year.

4.2.4. Residential wood combustion sources

Currently, the only federal regulation that applies to residential wood heating devices is the New Source Performance Standard (NSPS) for residential wood heaters. Adopted in 1988, the NSPS applies only to new units and has not been revised since its adoption. Efforts are underway involving a number of states to encourage the USEPA to update the 1988 NSPS in light of technology advances as well as broaden its applicability to a number of residential wood heating devices that are not covered by the 1988 NSPS.⁴² The types of devices currently exempted from NSPS include fireplaces, masonry heaters, pellet stoves, outdoor/indoor wood boilers, and outdoor/indoor wood furnaces. stopped

Wood combustion is an important emissions source of primary particulate matter in the MANE-VU region, as well as a source of VOCs (many of which are air toxics) contributing to SOA formation. Wood smoke also is a source of nitrogen oxides, which in addition to promoting SOAs also leads to nitrate aerosols, particularly in wintertime when nitrates are more thermally stable and residential wood combustion activity is at its highest. In Europe, emission standards for wood combustion devices go beyond particulate matter emissions, and require units to meet emission limits for CO, NO_x, PM, VOCs, and combustion efficiency. Combining a multipollutant approach with a thermal combustion efficiency standard can create significant co-benefits, including reduced emissions, ensuring no trade-offs among pollutants (e.g., decreasing CO emissions to very low levels while increasing NO_x emissions), and reduced fuel consumption. A suite of revised NSPS for a wider range of residential wood burning devices is an opportunity to incorporate advances in combustion technologies, improve efficiencies, and reduce a broad range of air pollutants affecting public health and visibility in the MANE-VU region.

4.2.5. Industrial, commercial, and institutional wood boilers

Total overall emissions from wood combustion in industrial, commercial, and institutional (ICI) boilers are small in comparison to residential wood combustion. There are efforts, however, to increase the use of ICI wood-fired boilers, such as at schools and hospitals. These settings, however, can expose sensitive populations to potential increases in particulate matter and other health damaging pollutants. There are some efforts underway at the federal and state levels to address emissions from this source category. For example, combined heat and power systems (CHP) (discussed below) offer the potential of using biomass in a more efficient and cleaner manner than burning in

⁴⁰ Mid-Atlantic Diesel Collaborative: <http://www.marama.org/diesel/>.

⁴¹ Northeast Diesel Collaborative: <http://www.northeastdiesel.org/>.

⁴² Letter to USEPA from Western States Air Resources Council (WESTAR) and Northeast States for Coordinated Air Use Management (NESCAUM), *re: New Source Performance Standard for Residential Wood Burning Devices* (August 3, 2012). Available at http://www.nescaum.org/documents/westar_nescaum-joint-nsps-res-wood-devices-letter-20120803.pdf/.

conventional combustion sources. Industrial, commercial, and institutional settings may be viable opportunities for expanding the use of CHP in the region.

4.3. Other strategies

4.3.1. Energy efficiency

The OTC is developing information on the potential for building energy efficiency improvements to reduce emissions, with a status report due in fall. The type of measures considered would be to replace existing windows with higher insulating windows and implement other energy efficiency measures at the same time a building is replacing its existing heating/ventilation/air conditioning (HVAC) system. Installing energy efficiency measures concurrently allows the purchase of a smaller replacement HVAC system at lower capital and operating costs. These types of retrofit projects have achieved profitable energy reductions with simple payback periods as short as three years. This would lead to pollution reductions through reduced energy consumption, and would occur year-round (i.e., not just during the ozone season). For example, in a May 2012 draft of its status report, the OTC estimates that deep energy retrofits of large office buildings in the Ozone Transport Region⁴³ (OTR) have potential to reduce NO_x by 36,000 tons annually.⁴⁴

4.3.2. Combined heat and power

MANE-VU has specifically identified combined heat and power (CHP) as an opportunity for reducing emissions contributing to regional haze and ozone formation, and has charged the MANE-VU Technical Support Committee with investigating the potential benefits of its expanded use in the region.⁴⁵ A CHP system, also called cogeneration, is an integrated energy system that generates electrical or mechanical power via fuel combustion (e.g., fossil fuels, biomass) while also capturing and using the waste thermal energy generated during combustion for heating purposes, thus operating at much higher efficiency levels than traditional combustion sources.

Well-designed CHP systems can reduce overall air pollutant emissions through several ways. The inherently higher efficiency of providing electricity and heat from the same combustion source reduces overall fuel consumption per given energy output, hence emissions as well. Where the generated electricity is used at the same location as the heat load, transmission and distribution losses from sending electricity over power lines are avoided. Improved reliability at the site can help avoid the use of emergency back-up generators (often uncontrolled and highly polluting diesel internal combustion engines)

⁴³ The Ozone Transport Region includes all of the MANE-VU region plus the northern Virginia counties in the Washington, DC area.

⁴⁴ Ozone Transport Commission (OTC) Energy Efficiency Workgroup. (2012). *Promoting Deep Energy Retrofits of Large Office Buildings To Reduce Nitrogen Oxide Emissions In the Ozone Transport Region*. OTC, Washington, DC, Status Report (May 8, 2012). Available at <http://www.otcair.org/upload/Documents/Meeting%20Materials/Draft%20Commercial%20Building%20Energy%20Efficiency%20Status%20Report%2005-08-12.pdf> (accessed September 14, 2012).

⁴⁵ MANE-VU. (2012). *Charge to the Technical Support Committee for Work in 2013*, adopted November 15, 2012. Available at <http://www.otcair.org/manevu/document.asp?fview=Formal%20Actions#> (accessed November 28, 2012).

during electrical grid outages. An added benefit is the economic savings from reduced energy costs due to higher CHP efficiency, which also provides a hedge against volatile fuel price swings.

Potential opportunities for applying CHP include:

- Industrial manufacturers – chemical, refining, ethanol, pulp and paper, food processing, glass manufacturing
- Institutions – colleges and universities, hospitals, prisons, military bases
- Commercial buildings – hotels and casinos, airports, high-tech campuses, large office buildings, nursing homes
- Municipal – district energy systems, wastewater treatment facilities, K-12 schools
- Residential – multi-family housing, planned communities

Site-specific factors that include matching the heat output with the thermal power load, value of reliability, and current energy costs typically determine the viability of CHP systems for a given application.⁴⁶

4.3.3. Ammonia

Ammonia is a key precursor in the formation of the aerosols ammonium sulfate and ammonium nitrate. While ammonia has a relatively short atmospheric lifetime of about 1 day, ammonium in particulate matter can have a lifetime of approximately 7-10 days, allowing for relatively long transport distances from source regions to downwind receptor areas.⁴⁷ On a national basis, the major sources of ammonia in the United States are livestock waste, fertilizer application, and soil emissions.⁴⁸ In urban areas, motor vehicles with 3-way catalytic converters may be a significant portion of the local ammonia inventory (>50 percent). Uncertainties in ammonia emission factors, spatial distributions, and seasonal variations can be large, particularly for area-wide sources such as livestock and fertilized croplands.⁴⁹

The potential of ammonia emission reductions to address fine particulate matter concentrations has received relatively little attention compared to emissions of sulfur oxides and nitrogen oxides. Several published modeling studies that include the eastern United States in the modeling domain suggest that ammonia reductions would have their greatest impacts during colder winter months.⁵⁰

⁴⁶ U.S. Environmental Protection Agency. (2012). *Combined Heat and Power Partnership: Basic Information*. Available at <http://www.epa.gov/chp/basic/index.html> (accessed October 26, 2012).

⁴⁷ Pinder, R.W., A.B. Gilliland, and R.L. Dennis. (2008). "Environmental impact of atmospheric NH₃ emissions under present and future conditions in the eastern United States." *Geophys. Res. Lett.*, **35**, L12808, doi:10.1029/2008GL033732.

⁴⁸ Davidson, C. et al. (2001). *Development of an Improved Ammonia Emissions Inventory for the United States*. Prepared for NESCAUM and MARAMA. Available by request from NESCAUM.

⁴⁹ Ellis, et al. 2009. "The influence of gas-particle partitioning and surface-atmosphere exchange on ammonia during BAQS-Met." *Atmos. Chem. Phys.* **11**, 133-145, doi:10.5194/acp-11-133-2011; Battye, W., V.P. Aneja, and P.A. Roelle. (2003). "Evaluation and improvement of ammonia emissions inventories." *Atmos. Environ.* **37**, 3873-3883, doi:10.1016/S1352-2310(03)00343-1.

⁵⁰ Makar, P.A. et al. (2009). "Modelling the impacts of ammonia emissions reductions on North American air quality." *Atmos. Chem. Phys.* **9**, 7183-7212; Pinder, R.W., A.B. Gilliland, and R.L. Dennis. (2008).

Information on the cost-effectiveness of ammonia controls relative to NO_x and sulfur dioxide reductions is limited, in part due to uncertainty in the ammonia emissions inventories and the limited control cost information for the major ammonia emission sources. The area-wide nature of agricultural ammonia emissions is also not necessarily amenable to typical air pollution control options. Available information on potential ammonia reduction options and estimated costs in Europe and the U.S. have been collected and tabulated by Pinder *et al.* (2007) and are reproduced in Table 4-2.⁵¹ These generally encompass manure management and fertilizer application practices in the agriculture sector.

Table 4-2. Costs of Ammonia Control Options (\$ per ton)

Ammonia Control Option	Cost (\$/ton)
Chemical additives to swine housing floor	70
Chemical additives to cattle housing floor	200
Cover broiler manure	30-300
Replace urea fertilizer with ammonium nitrate	500
Allow crust formation on lagoon surface	700
Immediate incorporation of applied manure	800
Chemical additives to poultry housing floor	900
Adapt poultry housing	2,500
Apply manure with trailing shoe	7,500
Adapt dairy housing	10,000
Rigid cover for pig manure stores	15,000
Belt drying layer (chicken) manure	20,000

From Pinder *et al.*, *Environ. Sci. Technol.*, 2007.

“Environmental impact of atmospheric NH₃ emissions under present and future conditions in the eastern United States.” *Geophys. Res. Lett.*, **35**, L12808, doi:10.1029/2008GL033732; Pinder, R.W. P.J. Adams, and S.N. Pandis. (2007). “Ammonia Emission Controls as a Cost-Effective Strategy for Reducing Atmospheric Particulate Matter in the Eastern United States.” **41**, *Environ. Sci. Technol.* 380-386, doi:10.1021/es060379a; Tsimpidi, A.P., V.A. Karydis, and S.N. Pandis. (2007). “Response of Inorganic Fine Particulate Matter to Emission Changes of Sulfur Dioxide and Ammonia: The Eastern United States as a Case Study.” **57**, 1489-1498, doi:10.3155/1047-3289.57.12.1489.

⁵¹ Pinder, R.W. P.J. Adams, and S.N. Pandis. (2007). “Ammonia Emission Controls as a Cost-Effective Strategy for Reducing Atmospheric Particulate Matter in the Eastern United States.” **41**, *Environ. Sci. Technol.* 380-386, doi:10.1021/es060379a.

5. SUMMARY

Efforts to reduce sulfate-forming air pollutant emissions are achieving success, as reflected by improved visibility and lower PM_{2.5} concentrations across the MANE-VU region during the past decade. However, in order to meet the national visibility goal of natural background conditions by 2064, the other components contributing to regional haze must also be addressed. In addition, locally elevated PM_{2.5} mass levels from wood combustion, car and truck exhaust, and other non-sulfate sources remain public health concerns. Based on monitoring data from the IMPROVE network, nitrate and carbonaceous PM_{2.5} constitute the next largest share of air pollution contributing to poor visibility and PM_{2.5} mass in the MANE-VU region.

While NOx reduction efforts can address the nitrate contribution, the recent scientific literature indicates reductions in NOx and primary carbon PM_{2.5} emissions can also have an important impact on reducing secondary organic aerosol formation. This is particularly noteworthy in the context “biogenic” secondary organic aerosols whose formation can be enhanced by controllable anthropogenic emissions. While these aerosols can be composed of organic components emitted by vegetation, the atmospheric chemistry leading to their formation can be greatly influenced by the presence of anthropogenic NOx and primary carbon PM_{2.5} emissions. As a result, strategies aimed at reducing nitrate and primary carbon aerosols can have the added benefit of reducing “biogenic” secondary organic aerosols as well.

As MANE-VU considers a long-term strategy beyond sulfate, it has a broad range of pollution reduction options for achieving air quality standards and visibility goals. At the federal level, tighter motor vehicle emission standards coupled with lower sulfur gasoline content can significantly reduce NOx and VOC emissions from mobile sources. Continued federal funding support for diesel equipment early retirement and retrofit measures would also contribute to continued progress in the region. States have been evaluating a number of source sector-specific NOx and VOC controls within the region with the goal of better public health protection, and these can also lead to regional visibility improvements as well.

In going beyond sulfates, the multiple constituents and complex atmospheric chemistry of the non-sulfate aerosols point towards multi-pollutant strategies to achieve future public health and visibility goals for the MANE-VU region. This dovetails with the trend in planning efforts across a broad spectrum of air, climate, and energy issues. Future visibility and PM_{2.5} NAAQS planning has the opportunity to take from these efforts, and contribute to them as well.