

White Paper: Approaches for Attaining the PM_{2.5} Daily Standard

The purpose of this paper is to summarize the results of several recent air quality studies and identify measures which will be effective in lowering PM_{2.5} daily concentrations in the Midwest. The air quality studies demonstrated that high daily PM_{2.5} concentrations occur year-round, but are more likely in the winter and summer months, and are associated with elevated concentrations of particulate sulfate (especially in the summer), particulate nitrate (in the winter), and organic carbon (OC). Effective control programs for these PM species include:

Regional reductions in sulfur dioxide (SO₂) emissions from EGUs and large non-EGUs

Reductions in ammonia (NH₃) emissions from agricultural operations, especially in winter

Regional reductions in oxides of nitrogen (NO_x) emission reductions

Urban-scale reductions in OC primary emissions from residential wood combustion and mobile sources, and VOC emissions from anthropogenic sources

Research Studies Considered

"A Conceptual Model of PM_{2.5} Episodes in the Midwest", LADCO, January 2009

Turner, J., "A Conceptual Model for Ambient Fine Particulate Matter over Southeast Michigan: High Concentration Days", Version 1.0, Final Report prepared for Southeast Michigan Council of Governments (October 2008)

Blanchard, C. and S. Tanenbaum, "Analysis of Inorganic Particulate Matter Formation in the Midwestern United States", Final Report, November 30, 2008.

Schauer, J.J., A. Rutter, D. Snyder, E. Stone, J. DeMinter, M. Mieritz, and B. Shelton, "LADCO Organic Molecular Marker Study Final Report", July 19, 2009, University of Wisconsin-Madison.

Sullivan, A.P., and J.L. Collett, "Biomass Burning Impacts on Air Quality in the Upper Midwest", Final Report, January 28, 2009, Colorado State University

Wade, K., "Analysis of High PM_{2.5} Days", Final Technical Memorandum, August 13, 2008, Sonoma Technology, Inc.

Wade, K., "Meteorological Analysis for of High PM_{2.5} Days", Final Letter Report, December 2, 2008, Sonoma Technology, Inc.

Brown, S., H. Hafner, P. Roberts, R. Sheesley, and J. Schauer, "Integration of Results for the Upper Midwest Urban Organics Study", Final Report, March 31, 2006, Sonoma Technology, Inc. and University of Wisconsin-Madison.

Lewandowski, M., M. Jaoui, J. Offenber, E. Edney, R. Sheesley, and J. Schauer, "Primary and Secondary Contributions to Ambient PM in the Midwestern United States", 2008, Environ. Sci. Technol. 42, 3303-3309.

Key Findings

- Nonattainment of the 24-hour PM_{2.5} NAAQS is a problem in the LADCO region, with 57 of 126 monitors exceeding the standard in 2005-2007, and 9 of 115 in 2006-2008. Most of the nonattainment monitors are in urban areas. On October 8, 2009, the U.S. Environmental Protection Agency (EPA) issued final area designations for the 24-hour NAAQS based on monitoring data from 2006 to 2008. EPA designated five areas in the LADCO region as nonattainment: Milwaukee, Detroit, Cleveland, Canton, and Steubenville.
- Since measurements of PM_{2.5} began in 1999, concentrations on the highest 90% days have fallen by about 0.5 ug/m³/year. Trends are consistently downward at all monitors with long term records. Meteorologically-adjusted trends (based on CART methodology), however, are relatively flat, suggesting that the actual trends are driven more by year-to-year variations in meteorology than by changes in emissions.
- Episodes of elevated concentrations generally occur across broad geographic areas, involving multiple cities and states. PM_{2.5} composition during an episode is similar across a city or affected area and is driven primarily by higher levels of: (a) ammonium sulfate during all seasons at all sites, (b) ammonium nitrate during the winter, especially at northern sites in the region, and (c) organic carbon during both summer and winter episodes.
- High daily concentrations depend on certain meteorological conditions: stagnant air masses associated with high pressure, slow wind speeds, high relative humidity, and southerly winds. The longer these conditions persist, the higher concentrations become, until a change in weather patterns brings less polluted air into the region.
- Local pollution sources can also be important contributors on high PM_{2.5} days in heavily industrial locations.

Recommended Control Measures

- Reductions in regional SO₂ emissions will be effective year-round. Based on the latest regional emissions inventory, the largest sources of SO₂ emissions are EGUs and non-EGUs.

Table 1. Annual SO₂ Emissions in LADCO Region (1000 TPY)

| | 2005 | 2012 | 2018 |
|--------------|--------------|--------------|--------------|
| Point-EGU | 2,826 (83%) | 1,665 (77%) | 1,468 (76%) |
| Point-NonEGU | 470 (14%) | 423 (20%) | 393 (20%) |
| Area | 47 (1%) | 44 (2%) | 42 (2%) |
| Nonroad | 61 (2%) | 16 (1%) | 11 (1%) |
| On-road | 20 (1%) | 5 (–) | 4 (–) |
| | 3,425 | 2,155 | 1,919 |

Given the spatial extent of elevated sulfate levels on high PM_{2.5} days, a regional control SO₂ program is necessary. This is best accomplished through a federal rule. On-going State Collaborative efforts are intended to assist EPA in developing a CAIR replacement rule (i.e., ensure that the geographic and source sector coverage, and the level of emission reduction, are sufficient to address interstate transport).

- Reductions in regional agricultural ammonia emissions will be most effective in the winter months and more effective at northern sites. Recent analysis of Midwest ammonia data shows that during winter months "...decreases in ammonia yield lower predicted PM_{2.5} than decreases in total nitrate at the majority of sites..." (Blanchard and Tanenbaum, 2008). (Further information will be forthcoming from analysis of the Winter Nitrate Study data.) The regional emissions inventory shows that ammonia emissions are dominated by agricultural operations year-round and fertilizer application in the spring and fall months. There are no federal programs to address ammonia emissions from agricultural operations. Possible control measures representing best management practices for ammonia are discussed in a white paper prepared by EC/R (2007).
- Reductions in regional NO_x emissions will also be effective in the winter months and more effective at northern sites (although, perhaps, not as effective as reductions in agricultural ammonia emissions). Based on the latest regional emissions inventory, the largest sources of NO_x emissions are EGUs, non-EGUs, and mobile sources (on-road and nonroad).

Table 2. Annual NO_x Emissions in LADCO Region (1000 TPY)

| | 2005 | 2012 | 2018 |
|--------------|--------------|--------------|--------------|
| Point-EGU | 788 (26%) | 651 (31%) | 638 (38%) |
| Point-NonEGU | 345 (11%) | 310 (15%) | 301 (18%) |
| Area | 163 (5%) | 164 (8%) | 159 (10%) |
| Nonroad | 583 (19%) | 386 (18%) | 274 (16%) |
| On-road | 1,188 (39%) | 597 (28%) | 299 (18%) |
| | 3,069 | 2,110 | 1,672 |

NO_x control options for stationary sources include combustion modifications, fuel treatment, and post-combustion controls for EGUs and non-EGUs (e.g., industrial, commercial, and institutional [ICI] boilers) – see MACTEC (2006). NO_x control options for mobile sources, including anti-idling, chip reflashing, diesel retrofits, identification/remediation of high emitting vehicles, and fleet/equipment modernization – see Environ (2007).

- Reductions in urban-scale organic carbon emissions will be effective year-round. A focus on urban-scale emissions is appropriate in light of a noticeable urban organic carbon increment compared to the regional background. Based on the Midwest source apportionment analyses (i.e., Schauer, et al, 2009; Sullivan and Collett, 2009; and

Brown, et al, 2006), contributors to organic carbon include biomass burning (about 10% in summer and 20-30% in winter), mobile sources (on the order of 25-30%), secondary organic aerosols (note, studies differ on the relative amounts attributable to anthropogenic and biogenic sources)¹, and, in industrialized areas, local point sources.

Based on these findings, possible control programs include:

- reductions in residential biomass combustion emissions from wood-burning stoves, fireplaces, and outdoor wood heaters, including a number of practices suggested by the EPA for reducing air emissions and improving efficiency <http://www.epa.gov/Woodstoves/>
- identifying and cleaning-up high emitting vehicles (e.g., poorly maintained on-road gasoline engines and off-road high emitting gasoline engines, such as construction equipment, load moving equipment, and two stroke engines used in yard maintenance). High emitting vehicle programs to consider are the Valley CAN pilot programs which attempt to identify and remediate high emitting vehicle problems. The pilot programs have been held in several cities across California since 2005 and have been successful in achieving cost effective reductions in HC, CO, and NO_x emissions (e.g., on the order of \$1,200 - \$2,100 per ton of CO, HC, and NO_x [combined] reduced).
- reductions in anthropogenic SOA due to toluene-like VOC precursors from motor vehicles, fuel spillage, and fugitive industrial solvent emissions
- reductions in primary OC emissions from local point sources, if relevant.

References (Candidate Control Strategies)

- MACTEC, "Identification and Evaluation of Candidate Control Measures, Phase II Final Report", June 2006
- Environ, "Evaluation of Candidate Mobile Source Control Measures for LADCO States in 2009 and 2012", March 21, 2007
- EC/R, "Interim White Paper: Midwest Regional Planning Organization, Source Category: Agricultural Ammonia Emissions", November 15, 2007
- U.S. Environmental Protection Agency, Cleaner Burning Woodstoves and Fireplaces, <http://www.epa.gov/Woodstoves/>

¹

| Study | Method | % Biogenic to SOA | Contribution (Summer) to Total OC |
|----------------------------|--|-------------------|-----------------------------------|
| Sullivan and Collett, 2009 | Radiocarbon Analysis | ---- | 35-50% |
| Lewandowski, et al, 2008 | GC-MS; monthly composite samples | 80% | 40% |
| Schauer, et al, 2009 | GC-MS solvent extraction; individual day samples | 33% | 15% |

- Valley CAN Reports:
 - The Valley CAN Tune In & Tune Up 2005 Program, Fresno, California, Sponsored by Valley CAN with Assistance from the Advanced Transportation Technology Center, Fresno City College, and California Bureau of Automotive Repair Breathe Easier Program, November 30, 2006
 - The Valley CAN Tune In & Tune Up 2006 Program, Bakersfield, California, Sponsored by Valley CAN with Assistance from The Advanced Transportation Technology and Energy Initiative Center, Bakersfield College, and California Bureau of Automotive Repair Breathe Easier Program, September 21, 2007
 - The Valley CAN Tune In & Tune Up 2006 Program, Stockton, California, Sponsored by Valley CAN with Assistance from The Advanced Transportation Technology and Energy Initiative Center, San Joaquin Delta College, and California Bureau of Automotive Repair Breathe Easier Program, January 4, 2008
 - The Valley Clean Air Now (CAN) Tune In & Tune Up 2007 Program, Arvin, California, Sponsored by Valley CAN with Assistance from The Advanced Transportation Technology and Energy Initiative, Arvin High School, and The California Bureau of Automotive Repair, July 10, 2008
 - The Valley Clean Air Now (CAN) Tune In & Tune Program Events at Visalia, Merced, Fresno, and Modesto, California, 2006-2007. Sponsored by Valley Clean Air Now with assistance from The Advanced Transportation Technology and Energy Initiative, February 27, 2009
 - Valley Clean Air Now (Valley CAN) Tune In & Tune Up Program, Parlier, California. Sponsored by Valley Clean Air Now with assistance from The Advanced Transportation Technology and Energy Initiative, The Kenneth L. Maddy Institute at California State University, Fresno, The California Department of Consumer Affairs, Reedley College, and Parlier High School, July 24, 2009