On the Air

Technical Notes on Important Air Quality Issues

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Fine Particle Levels in the Great Smokies: Sorting Out All the Sources

Science: Understanding sources of aerosols requires a complex analysis of data obtained by continuous monitoring of a "background" site. After sulfates, carbon-containing constituents comprise the largest fraction of fine particle mass in the southeastern U.S. The Tennessee Valley Authority's research at Look Rock, on the western edge of the Great Smoky Mountains National Park, continues to make progress in its investigation of the sources of carbon-based particles. The research serves as a basis for suggesting appropriate environmental management strategies.

Policy: Knowledge of all the sources of fine particles is essential to the development of approaches that are effective in the achievement of the health-based fine particle National Ambient Air Quality Standard and in the improvement of visibility in Class I areas. Background monitoring helps evaluate whether control strategies are effective in meeting regulatory goals.

Rural background sampling sites, such as the Look Rock site near the Great Smoky Mountains National Park (GSMNP) help sort out regional sources of pollutants, such as ozone and fine particles. Samples from such remote background sites are examined to determine the natural levels of pollutants in air without the influence of nearby sources. Background data and its implications about sources should prove useful in the development of control strategies.

In 1999, the Tennessee Valley Authority (TVA) expanded its traditional filter sampling methods for continuous monitoring of fine particulate matter ($PM_{2.5}$) at the Look Rock site to include natural organic contributions from trees and other vegetation. The short-time resolution methods used at Look Rock, combined with meteorological measurements and source profile evaluations, are improving our ability to understand all the sources that contribute to airborne fine particles and identify approaches to reduce them.

What sources of fine particles and their precursors can actually be controlled?
The fundamental question of how best to manage $PM_{2.5}$ levels is being revisited as a result of the recent finding that carbon is a major component of fine particle composition. Controls on emissions of sulfur dioxide (SO_2) from coal-fired power plants mandated by the Clean Air Act have resulted in some reductions in secondary fine

particle sulfate in the eastern U.S., including the Tennessee Valley. Reductions in organic and elemental carbon levels in fine particles are also needed, and may be more important from the standpoint of health effects. These reductions should be at least partially addressed by recently finalized rules on highway and off-road diesel vehicles and other pending regulations. However, the sources of elemental and organic carbon-based particles are a complex mix of anthropogenic and natural biogenic emitters. TVA's fine particle measurement program over the past few years at Look Rock has studied the composition and properties of these carbonaceous particles to gain insight into what steps can be taken to control sources to reduce this major fraction of the $PM_{2.5}$ mass.

Extensive monitoring of the composition of fine particles in the southeastern U.S. conducted since formulation of the 1997 $PM_{2.5}$ National Ambient Air Quality Standard (NAAQS) has shown that fine particles consist chiefly of sulfates and of organic and elemental carbon, along with small amounts of nitrates, trace elements, and bound water (Figure 1).

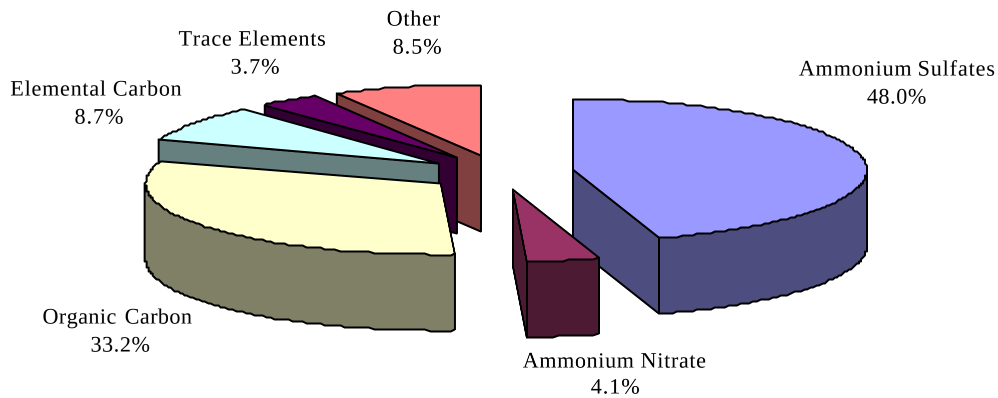

Figure 1. Average fine particle composition at Look Rock across all seasons (2001 TVA data).

Trace elements and elemental carbon mostly derive from direct emissions from industrial processes and natural and human-caused combustion sources. Sulfate and nitrate particles are secondary products formed by atmospheric conversion of gaseous SO_2 and nitrogen oxides (NO_x) emissions that chiefly come from power plants and vehicles.

We know how to reduce primary emissions from combustion processes that produce organic (carbon) aerosols by controlling their sources—diesel fuel combustion, wood burning, etc. Likewise, sources of directly emitted elemental and metallic aerosols from industrial sources and utilities can be directly controlled. Secondary sulfate and nitrate levels in the atmosphere are being reduced by controlling the emission of their gaseous precursors, SO_2 and NO_x, from coal-fired power plants.

In contrast, organic carbon constituents of aerosols are a combination of directly emitted species (primary) and those formed by atmospheric conversion (secondary). This combination of primary and secondary generation complicates determination of effective, specific control strategies. Central questions being addressed by monitoring at the Look Rock site and in the Smokies include: What is the average chemical composition of fine particles at the Look Rock site and the Smokies? How does it vary by season and why?

Due to extensive efforts by the National Park Service (NPS) and TVA at Look Rock, Cove Mountain, and other sites, knowledge of aerosol chemical composition in the GSMNP is considerably more extensive now than it was a decade ago. As shown in Figure 1, on an annual average basis, ammonium sulfate is the most abundant $PM_{2.5}$ fraction at Look Rock. However the carbon-containing components, both organic and elemental, make up most of the remainder. These annual averages mask substantial seasonal variation in $PM_{2.5}$ composition. When source and season are examined, sulfate levels are higher in the summer and lower at other times of the year (Figure 2). Compared to rural or "background" levels, urban levels of $PM_{2.5}$ mass are higher in autumn largely because there are additional sources of organic aerosols, such as wood-burning, during this period as well as during winter (winter data not shown).

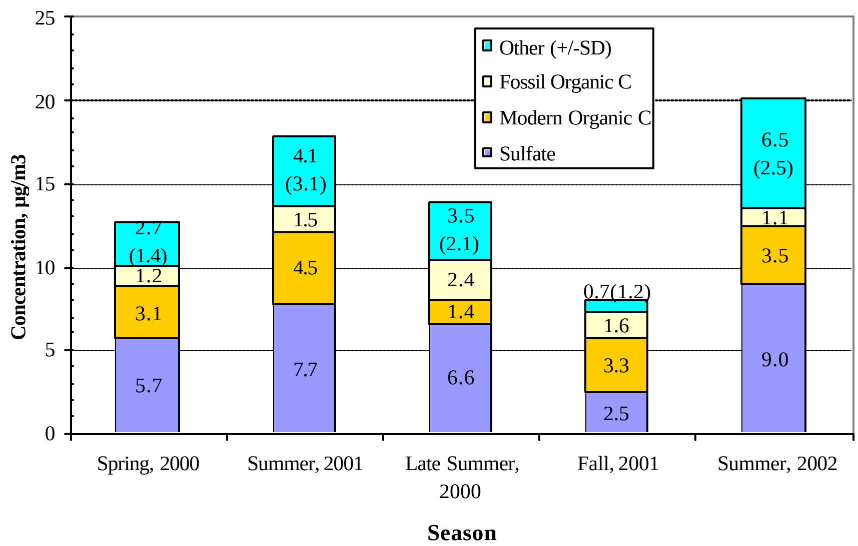

Figure 2. Average fine particle composition at Look Rock by source and season (based on carbon-14 measurement).

How are Look Rock data on daily and hourly aerosol composition being used?
With the short-time resolution provided by newly-developed instruments, we are beginning to understand how the concentrations of different fractions of $PM_{2.5}$ vary during the day, as well as seasonally. By comparing averaged hourly data and daily results with predictions by air quality computer models, the ability to model hour-to-hour variations in pollutant concentrations can be tested. Preliminary efforts indicate organic carbon aerosols are difficult to model accurately. Thus, TVA has taken the lead in attempting to pinpoint the sources of organic aerosols that make up this large fraction of regional $PM_{2.5}$ mass. Methods include measurement of carbon-14 (radiocarbon) and other tracers to help identify specific aerosol constituents and their sources (e.g., wood smoke, biomass burning). Carbon-14 concentration indicates what fraction of the organic aerosol is from fossil or "old" carbon sources (e.g., emissions from vehicles) and what fraction is from modern carbon (e.g., wood-burning). Finding tracers that are emitted in specific ratios by individual sources allows us to determine which fossil and biogenic carbon emissions are due to human activities and potentially controllable.

Some counties in the vicinity of Look Rock are currently being considered for nonattainment designation under the $PM_{2.5}$ NAAQS. As states proceed in planning measures to bring such areas into attainment with the annual fine particulate standard, the full body of sources of $PM_{2.5}$ will need to be evaluated.

How will VISTAS and other RPOs benefit from the hourly data from Look Rock and other focus sites?
The Visibility Improvement State and Tribal Association of the Southeast (VISTAS) and other Regional Planning Organizations (RPOs) deal with haze regulations. VISTAS is responsible for evaluating options for meeting regional haze regulations in the southeastern U.S. One goal is to provide field data, acquired by TVA and NPS at Look Rock and other sites, to modelers in order to evaluate whether existing computer models

can reproduce daily patterns of aerosol mass and composition. Model testing is important since current models for $PM_{2.5}$ prediction are still in an early stage of development. Determining natural organic aerosol levels is also important since the ultimate goal of the haze regulations is to return to "natural" conditions by 2064. Thus, quantifying and understanding "background visibility" or "natural conditions" is an important consideration for VISTAS and other RPOs that deal with the haze regulations. Having a year-round, hourly-averaged data base for aerosol constituents will also facilitate the determination of the aerosol composition on both the clearest and haziest days in Class 1 areas of the Appalachians, another component of haze regulations.

What is expected to result from a new VISTAS initiative on sources of organic aerosols?

A new VISTAS initiative will extend the particle characterization work done by TVA at Look Rock and elsewhere. Selected aerosol samples, collected by sampling equipment at the VISTAS focus sites, will be analyzed to determine the sources of aerosol carbon using organic tracer measurements. Major progress can then be expected in unraveling the organic aerosol "mystery" by identifying contributions from primary and secondary, biogenic and anthropogenic, controllable and "natural" components. Control strategy development requires this level of detail to reliably achieve reduced regional aerosol loadings in the most cost-effective manner.

Conclusions

Through improved knowledge of the carbon-based $PM_{2.5}$ constituents of background air, TVA can aid federal and state regulators in reducing $PM_{2.5}$ levels in the region. This knowledge is essential to the identification of the most effective approaches for meeting health-based $PM_{2.5}$ NAAQS and improving visibility in Class I areas. Technique development also allows the air quality community to evaluate more reliably whether control strategies are actually proving effective in meeting various regulatory goals.

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