

2nd Quarter 2003**The IMPROVE Newsletter**

Volume 12 / Number 2

Monitoring update

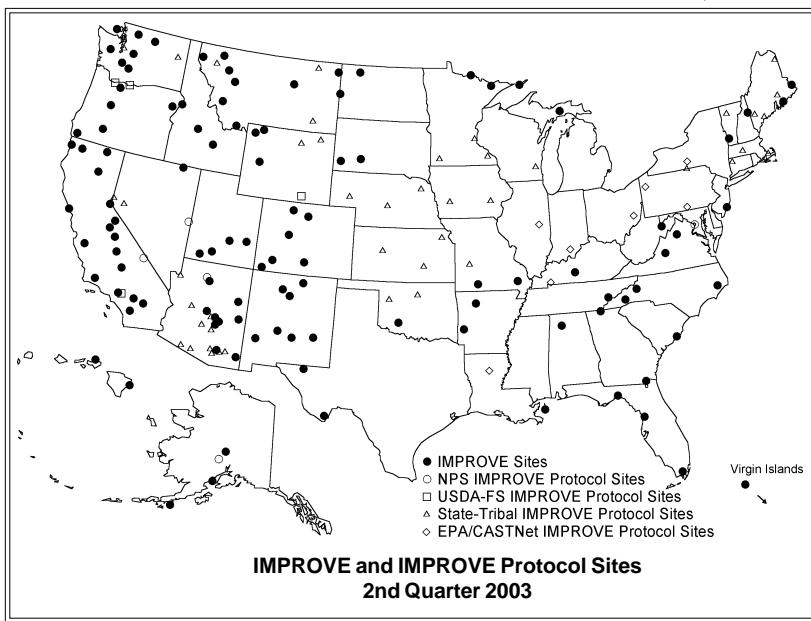
Network operation status

The IMPROVE aerosol monitoring network consisted of 110 aerosol samplers during 2nd Quarter 2003 (April, May, and June). Additional instrumentation that operates according to IMPROVE protocol in support of the program included:

- 53 aerosol samplers
- 22 transmissometers
- 40 nephelometers
- 15 film or digital camera systems
- 11 Web camera systems
- 3 interpretive displays

Federal land managers, states, tribes, and other agencies operate supporting instrumentation at monitoring sites as presented in the map below. Preliminary data collection statistics for the quarter are:

- Aerosol (channel A only) 94% collection
- Aerosol (all modules) 92% completeness
- Optical (transmissometer) 94% collection
- Optical (nephelometer) 98% collection
- Scene (photographic) 96% collection



Feature Article: Yosemite study shows importance of smoke particles, Page 4

Several new monitoring sites were installed during the 2nd quarter. One aerosol sampler was installed at the Walker River Reservation, NV, and will operate according to IMPROVE Protocol. One transmissometer was installed at Mesa, AZ, and nephelometers were installed at National Capital-Central, DC; St. Paul, MN; Upper Buffalo Wilderness, AR; Cedar Bluff State Park, KS; Wichita Mountains National Wildlife Refuge, OK; and Organ Pipe, Children's Park, Queen Valley, Estrella Mountain Park, Vehicle Emissions Station, and Dysart Road, all in Arizona.

Web cameras were installed at National Capital-Central, DC; St. Paul, MN; Mount Hood, OR; Vancouver, WA; and Camelback Mountain, South Mountain, Superstition Mountains, Estrella Mountain Park, and White Tank Mountains, all in Arizona.

Data availability status

Data are available on the IMPROVE Web site, at <http://vista.cira.colostate.edu/improve/Data/data.htm>. IMPROVE and other haze related data are also available on the VIEWS Web site, at <http://views.vista.cira.colostate.edu>. Aerosol data are available through November 2002. Transmissometer data are available through December 2001 and nephelometer data are available through March 2003.

Photographic slides and digital images are archived but are not routinely analyzed or reported. Complete photographic archives and slide spectrums (if completed) are available at Air Resource Specialists, Inc. Slide spectrums are also available on the IMPROVE Web site, under *Data*. Near real-time digital images from National Park Service Web camera sites can be viewed at <http://www2.nature.nps.gov/ard/cams/index.htm>. USDA Forest Service camera images from IMPROVE sites can be viewed at <http://www.fsvisimages.com>.

Monitoring update continued on page 3....

Visibility news

EPA changes regional haze rule to reduce sulfur dioxide in western United States

The Environmental Protection Agency (EPA) has amended its regional haze rule to aid states and tribes in the western United States improve visibility in the 16 Class I areas on the Colorado Plateau. New provisions incorporated into the rule set regional milestones for reducing sulfur dioxide emissions, a key component of regional haze. The milestones are part of a program for visibility in nine western states and eligible Indian tribes within the same geographic area.

The new provisions were first proposed to the EPA in September 2000 by the Western Regional Air Partnership (WRAP), which includes representatives of western states, tribes, and federal agencies. The proposal is also known as the Annex to the 1996 Report of the Grand Canyon Visibility Transport Commission (GCVTC). It includes recommended regional emissions reduction milestones for sulfur dioxide, and a description of a "backstop" emissions trading program for states and tribes in the Colorado Plateau area. If total regional emissions exceeds the annual milestone, the backstop trading program will initiate, ensuring emission reductions that will meet the reduction milestones.

Sulfur dioxide sources in the Colorado Plateau region currently emit approximately 650,000 tons per year. The new changes to the regional haze rule will reduce emissions by more than one-fourth by 2018. A copy of the rule may be downloaded from EPA's Web site at <http://www.epa.gov/ttn/oarpg> under Recent Actions.

For more information contact Tim Smith at EPA's Office of Air Quality Planning & Standards. Telephone: 919/541-4718. Fax: 919/541-5489. E-mail: smith.tim@epa.gov.

EPA proposes correction to mobile source provisions in regional haze rule

The Environmental Protection Agency (EPA) has proposed to correct mobile source provisions in Section 309 of the regional haze rule. The Western Regional Air Partnership (WRAP) recommended the corrections, which address emissions projections for mobile sources that were not addressed at the time the regional haze rule was published in 1999. If EPA receives no adverse comments, the final rule is expected to become effective September 2, 2003.

Section 309 requires states to establish mobile source emissions budgets for each area that significantly contributes to visibility impairment in any of the 16 Class I areas on the Colorado Plateau. In 1996, mobile source emissions were projected to be lowest in 2005 and to rise over the course of the first regional

haze planning period (2003-2018). Section 309 required mobile source emissions budgets to be set using the lowest projected level as a planning objective and performance indicator.

Since the budget recommendations were first developed, new developments have caused mobile source emissions projections to change significantly. New mobile source emissions models and new federal engine and fuel standards were adopted by EPA. As a result of these new models and standards, WRAP now projects a significant decline in mobile source emissions during 2003-2018, particularly from on-road mobile sources.

The revisions would change Section 309 to reflect emissions budgets that show a continuous decline over the planning period, as the new planning objective and performance indicator.

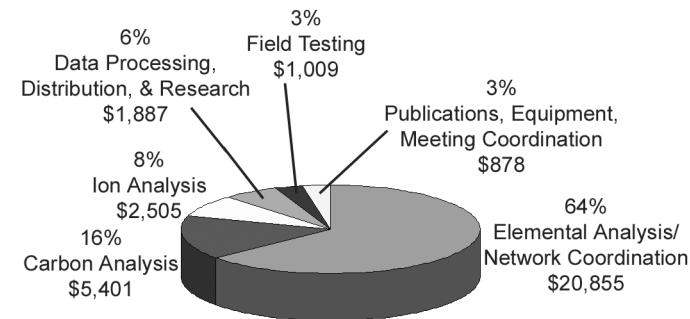
For more information contact Patrick Cummins at the Western Governor's Association. Telephone: 970/884-4770. E-mail: pcummins@westgov.org.

FY2003 IMPROVE aerosol monitoring budget

The total IMPROVE aerosol monitoring program budget for Fiscal Year 2003 (October 1, 2002 through September 30, 2003) is \$6,961,849. Of this amount, \$5,684,083 (81.6%) is EPA's contribution, used exclusively for the components listed in the graphic below.

The cost to operate and maintain an IMPROVE aerosol monitoring site for one year is \$32,535 (excluding operator salary and procurement of new or replacement equipment). Both the IMPROVE aerosol monitoring program and federal land managers (FLMs) fund the program. Operator salaries, optical and scene monitoring, and Web cameras are not funded by the IMPROVE aerosol monitoring program.

2003 IMPROVE Aerosol Monitoring Program Costs by Component



For more information contact David Maxwell at the National Park Service Air Resources Division. Telephone: 303/969-2810. Fax: 303/969-2822. E-mail: david_maxwell@nps.gov.

Visibility news continued on page 7....

Monitoring update *continued from page 1*

Outstanding sites

Data collection begins with those who operate, service, and maintain monitoring instrumentation. IMPROVE managers and contractors thank all site operators, for their efforts in operating the IMPROVE and IMPROVE Protocol networks. Sites that achieved 100% data collection for 2nd Quarter 2003 are:



Aerosol

Addison Pinnacle
 Arendtsville
 Badlands
 Boundary Waters
 Breton
 Bridger
 Brigantine
 Bryce Canyon
 Cabinet Mountains
 Caney Creek
 Cape Cod
 Casco Bay
 Cedar Bluff
 Chiricahua
 Cloud Peak
 Columbia Gorge East
 Columbia Gorge West
 Connecticut Hill
 Crescent Lake
 Denali
 El Dorado Springs
 Ellis
 Flathead
 Fort Peck
 Glacier
 Grand Canyon
 Great Gulf
 Great River Bluffs
 Great Sand Dunes
 Hawaii Volcanoes
 Hercules-Glades
 Hillside
 Hoover
 Ike's Backbone
 Isle Royale
 James River
 Jarbidge
 Joshua Tree
 Kalmiopsis
 Lava Beds
 Linville Gorge
 Livonia
 Lostwood

Lye Brook
 Meadview
 Mohawk Mountain
 Moosehorn
 Mount Rainier
 Okefenokee
 Old Town
 Olympic
 Organ Pipe
 Petrified Forest
 Pinnacles
 Presque Isle
 Seattle
 Quabbin Reservoir
 Quaker City
 Queen Valley
 Rocky Mountain
 Sac and Fox
 Saguaro West
 San Gabriel
 San Gorgonio
 Shenandoah
 Sikes
 Simeonof
 Snoqualmie Pass
 Starkey
 Sycamore Canyon
 Tallgrass
 Three Sisters
 Trapper Creek-Denali
 Trinity
 Upper Buffalo
 Viking Lake
 Virgin Islands
 Voyageurs
 Walker River
 Washington DC
 White Pass
 White River
 Wind Cave
 Yosemite
 Zion

Transmissometer

Grand Canyon (In-Canyon)
 Grand Canyon (South Rim)

Rocky Mountain

Nephelometer

Acadia
 Big Bend
 Mammoth Cave
 Mount Rainier

Quaker City
 Seney
 Virgin Islands

Photographic

Bryce Canyon
 Grand Canyon

Red Rock Lakes

Operators of distinction

Denali National Park and Preserve has been an IMPROVE site since 1986, when stacked filter units collected preliminary aerosol data before the network was officially inaugurated in 1988. Until network expansion occurred in 2001, Denali was the only long-term IMPROVE site in Alaska. Currently there are three other sites in the state, with a fourth station slated for installation this fall near Western Arctic National Parklands.

Andrea Blakesley, the primary site operator at Denali, worked with National Park Service (NPS) scientists and managers in Alaska and nationwide to ensure that an arctic site was added to the IMPROVE and NPS air quality monitoring networks. Along with the NPS Regional Air Quality Coordinator, Bud Rice, she promotes awareness of arctic and subarctic airborne contaminant issues, including the accumulation of persistent organic pollutants and other toxic airborne contaminants in high latitude ecosystems.

When Andrea first began operating the IMPROVE instruments in the early 90's, the common assumption was that the remote and rugged Denali wilderness was clean and pristine, virtually untouched by human pollution. But in looking more closely at the data, she saw an interesting pattern emerge. While the air in Denali is in fact exceptionally clean, each winter and spring there are spikes of contaminants that cannot be explained by local sources alone. A literature search confirmed that there are two transport pathways that bring industrial and agricultural contaminants into the arctic and subarctic throughout the circumpolar north, particularly from source areas in Europe and Asia. The seasonal patterns of transport match what is observed in Denali.

Like many before her, Andrea continues to be inspired by the beauty and wildness of Denali's intact ecosystems, which will be influenced over time as much by human development on other continents as by our national environmental protection laws.



Because of a historical near miss, Denali National Park and Preserve is the only Class I national park in Alaska. If the 1977 Clean Air Act amendments had occurred three years after instead of three years before legislation was passed to expand park and refuge lands in Alaska, Denali would have been one of eight mandatory Class I parks in the state.

Feature article

Aerosol properties during the Yosemite Aerosol Characterization Study of 2002

(by C. Carrico, J. Carrillo, T. Lee, J. Collett, S. Kreidenweis, P. Herckes, G. Engling, G. McMeeking, D. Day, W. Malm, T. Cahill, and G. Bench)

Introduction

Effective air quality and forest management strategies must consider EPA's Regional Haze Rule requiring Class I areas to attain natural background visual air quality by the middle of this century¹. The role of smoke from prescribed burns and wildfires in visibility degradation is poorly characterized as a result of few comprehensive smoke measurement campaigns. The National Park Service sponsored a study in Summer 2002 to improve understanding of aerosols, and in particular smoke impact on visibility in western U.S. parks.

Long-term IMPROVE measurements have suggested that national parks in the west are particularly impacted by forest fire smoke². Inland national parks of California such as Sequoia, Joshua Tree, and Yosemite National Parks are typically the most air quality impacted parks in the western U.S., particularly in summer^{2,3}. In addition to frequent smoke impacts, Yosemite is often downwind of population/agricultural centers of central California including the San Francisco Bay area and the San Joaquin Valley. The ozone concentration in Yosemite often exceeds the California standard of 90 ppb in the summer including the summer of 2002. As a result, Yosemite has demonstrated severe damage to flora from ozone³. Historically at Yosemite, the total light extinction coefficient peaks in the summer (the average summer transmissometer measurement from 1988-1997 was 70 Mm^{-1}) with a contribution approaching half from carbonaceous aerosol².

Visibility is critically dependent on aerosol phase water, and the precise hygroscopic (water uptake) behavior of particulate organic material (POM) is subject to considerable uncertainty^{4,5}. Moreover, as a result of the species that comprise it, fresh versus aged POM likely behave differently as do POM from biomass burning versus POM from an urban-industrial plume⁶. At high relative humidity, POM has been assumed to uptake no water² to being as hygroscopic as ammonium sulfate. Thus, the Yosemite Aerosol Characterization Study focused on three points:

- 1) to examine high time resolution aerosol chemistry measurements,
- 2) to investigate hygroscopic properties of POM dominated aerosol, and
- 3) to examine various tracers for wood smoke impacts.

Experimental methods

The Yosemite Aerosol Characterization Study involved aerosol and gas measurements made atop Turtleback Dome, and collocated with standard IMPROVE measurements. Researchers from Colorado State University performed the study in Yosemite National Park from July 15 to September 4, 2002. Measurements included physical, chemical, optical, and hygroscopic measurements.

High time resolution aerosol chemistry measurements

High time resolution chemical characterization included measurement of soluble PM_{2.5} ions with a Particle into Liquid Sampler (PILS) with 15-minute time resolution. A 2-channel aethalometer measured absorption by the collected aerosol at 350 and 880 nm with 5-minute time resolution. While the 880 nm channel is sensitive mainly to black carbon (BC), the 350 nm channel also responds to absorption by certain organic compounds, and a separation in the two channels indicates presence of diesel exhaust or wood smoke.

Hygroscopic properties of POM dominated aerosol

Hygroscopic measurements included size and light scattering growth factors using a humidified tandem differential mobility analyzer (HTDMA) and a humidified nephelometry system, respectively.

Tracers for wood smoke impacts

PM_{2.5} total carbon (TC) was measured hourly with a Rupprecht and Patashnick 5400 Carbon Monitor. Organic carbon (OC) was derived as the difference between total and black carbon as well as from TOR analysis of 24-hour IMPROVE filters. OC was multiplied by 1.6 giving particulate organic material (POM) to account for additional elements present in the organic material. Carbon isotope measurements were used to determine relative contributions of fossil vs. biogenic carbon sources. A 2-component model determined the fraction of carbon on each filter derived from biogenic sources, based on the measured ¹⁴C/C ratio. Organic speciation using solvent extraction and quantification by Gas-Chromatography coupled to Mass Spectrometry (GC-MS) was used to further characterize POM.

Results and discussion

Summer 2002 was one of the most active fire seasons in recent years in the west, and several smoke episodes were sampled as a result of hundreds of thousands of acres that burned in Oregon and California. During much of the summer, a regional haze engulfed the San Joaquin Valley with 11 "hazardous air

days" and 14 days when schools were advised to cancel outdoor activities. This compares with 1 hazardous air day in 2001 and is the highest number of severely polluted days since 1998⁷.

PM_{2.5} was dominated by particulate organic material (82%) with 24-hour average POM from IMPROVE TOR analysis varying from 1 to 18 $\mu\text{g}/\text{m}^3$ (Figures 1 and 2). Historically, average PM_{2.5} mass concentration in Yosemite during the summer was 6 $\mu\text{g}/\text{m}^3$ with a 50% contribution from POM as indicated from IMPROVE measurements³, demonstrating Summer 2002 to be a heavily impacted period. Based on 24-hour IMPROVE filters analyzed with TOR analysis, the POM/Elemental Carbon (EC) ratio at Yosemite was 15 on average, and reached 30, quite high compared with historical values and consistent with smoke. The POM/EC ratio during the week of the most smoke impacted period shown in Figure 2 averaged 20, similar to values found elsewhere in smoke plumes⁸ and much higher than those observed in polluted urban areas.

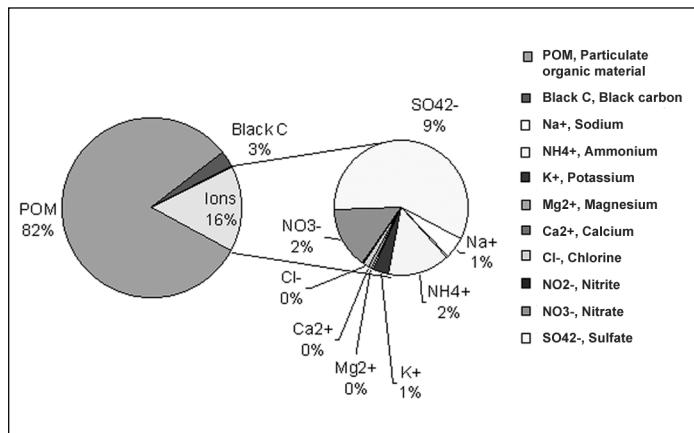


Figure 1. Average PM_{2.5} aerosol composition during the Yosemite Study in Summer 2002 (not including minor contributions from mineral dust species).

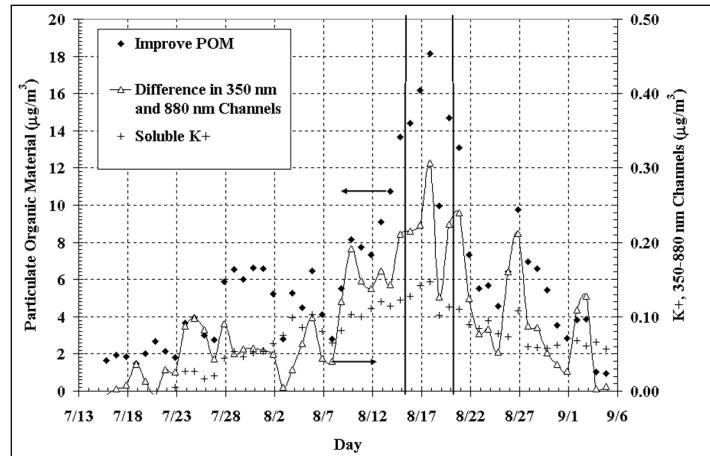


Figure 2. Mid-August was a strongly smoke impacted period as seen in 24-hour average POM concentrations from IMPROVE filters, a difference in 350 nm and 880 nm channels from the aethalometer measurement, and K⁺ concentration from PILS measurements.

The week between approximately August 13-21 featured the most substantial smoke impact from multiple indicators (Figure 2). This period featured the highest POM concentrations, the largest divergence of 350 nm and 880 nm channel absorption from the aethalometer, and the highest soluble potassium, likewise a biomass smoke indicator⁹. The modern carbon fraction exhibits a similar profile peaking above 95% in mid-August, though it never is less than 70%. Also evident is the predominance of accumulation mode particles (diameter ~ 500 nm) during this period. Likewise, transport patterns indicated the influence of large wildfires in Oregon as shown by air mass backtrajectories computed with the NOAA Hysplit model and MODIS (<http://visibleearth.nasa.gov>) satellite imagery.

Carbon isotopic analysis from daily filters showed that modern (rather than fossil) carbon comprised 73% to 95% of the PM_{2.5} carbon (Figure 3). Nearly all variability in total carbon mass was from modern carbon (from either biogenic sources or wood smoke). Organic speciation showed that vehicular emission tracers (hopanes and steranes) were detected at low concentrations suggesting less than 10% contribution to average POM. Concentrations of pinene oxidation products, including pinic and pinonic acids were high, accounting for several percent of total OC on average, and suggesting the contribution of secondary organic aerosol from biogenic sources. Certain periods, particularly during mid-August, showed a clear influence from wildfires, illustrated by high concentrations of wood smoke markers (resin acids, anhydrosugars, methoxyphenols).

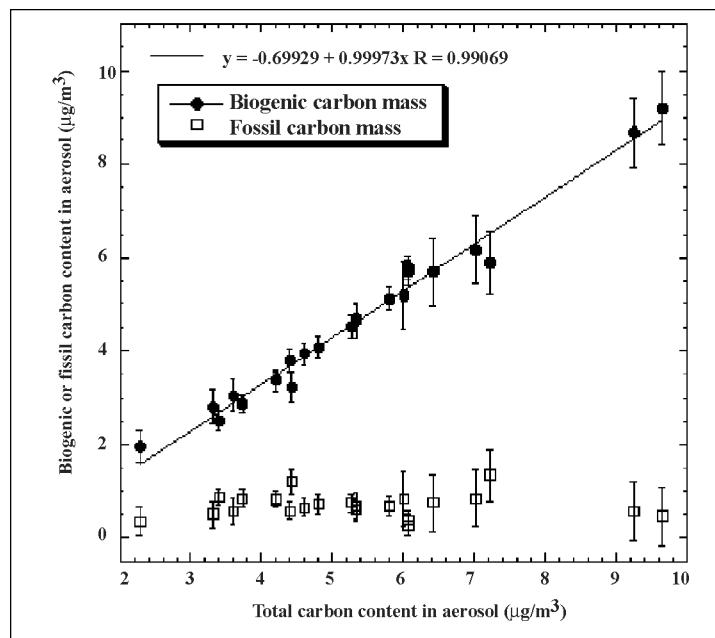


Figure 3. Relationship of biogenic carbon mass and fossil carbon mass to total PM_{2.5} carbon based on carbon isotope measurements.

High time resolution measurements showed strong diurnal patterns associated with the mountain-valley thermal system, which imposed distinct, though sometimes differing, diurnal cycles on aerosol species. Generally though, upslope westerly flow each morning initiated transport of a quiescent haze layer from the valleys below. The pronounced diurnal pattern in OC concentrations including afternoon peaks also suggested a natural biogenic contribution. The ionic fraction of the aerosol was typically dominated by ammoniated sulfate (Figure 1). Close tracking of NO_3^- and Na^+ concentrations provided strong evidence of the upwind reaction of nitric acid with sea salt before arrival at Yosemite. A diurnal pattern in aerosol size distribution was also evident with concentrations of accumulation mode particulates often increasing beginning in the morning, and an ultrafine mode often appearing in the afternoon, perhaps resulting from nucleation events.

As a result of the dominant POM contribution, aerosol hygroscopic properties differed greatly from typical marine or polluted aerosols that are usually dominated by ionic components such as Na^+ and Cl^- for the former, and NH_4^+ , SO_4^{2-} , and NO_3^- for the latter. Though limited hygroscopic growth was typically observed for $\text{RH} < 70\%$, beyond $\text{RH} = 80\%$, growth was much weaker than typical ionic species.

Particles of 200 nm grew by ratios of 1.14 and 1.29 at $\text{RH} = 80\%$ and 90%, respectively (compared with ~1.5 and 1.7 for pure $(\text{NH}_4)_2\text{SO}_4$ particles). A strong inverse relationship between carbon content and hygroscopicity was particularly evident with 200 nm particles ($R^2 = 0.72$ using an exponential decay fit) with growth factor at $\text{RH} = 80\%$ approaching 1.1 as the POM/ionic mass ratio exceeded 10 (Figure 4). Light scattering measurements showed yet lower growth due to the predominance of smoke in the optically important range. The

results showed that hygroscopic growth of smoke particles, though not hygrophobic, is considerably weaker than typical salt particles such as sulfates, nitrates and sea salt.

Conclusions

During Summer 2002, air quality in the San Joaquin Valley was the worst in several years and much of this regional haze reached the sampling site at Turtleback Dome in Yosemite National Park. Fine aerosol mass at the site was dominated by organic carbon, and under these hazy conditions, the predominance of biogenic sources was evident. Multiple indicators including carbon isotopic analysis, trace species, satellite imagery, and air mass backtrajectories indicated a strong impact of wildfire smoke in addition to other biogenic sources. The smoke impact was pervasive throughout the range of aerosol chemical, physical, optical, and hygroscopic properties measured at Yosemite.

Acknowledgments

This research was supported by the National Park Service. The authors acknowledge the vast assistance of Air Resource Specialists, Inc., and Russell Galipeau, Katy Warner, and the resource managers at Yosemite National Park.

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For more information contact Kip Carrico at Colorado State University. Telephone: 970/491-8667. Fax: 970/491-8483. E-mail: carrico@lamar.colostate.edu.

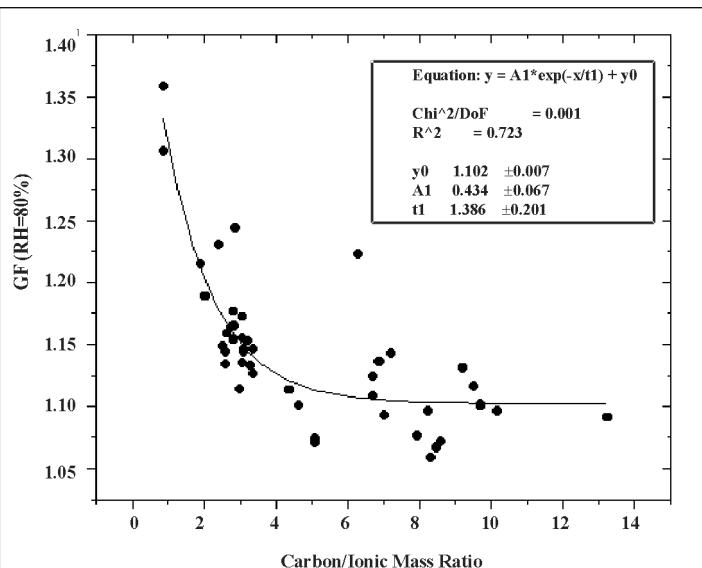


Figure 4. Relationship between carbon content and hygroscopic growth given as the carbon/ion mass ratio in $\text{PM}_{2.5}$ vs. diameter growth factor at $\text{RH} = 80\%$ of 200 nm dry particles (GF (RH=80%)).

Visibility news *continued from page 2*

IMPROVE committee meets in Tucson

The IMPROVE Steering Committee met in Tucson, Arizona, in June, to discuss current operations and research issues of interest to the program. Other major discussions included special studies, network comparisons, operator support, and technology innovations. Steering committee members, program contractors, and attendees from regional planning organizations also toured the IMPROVE and IMPROVE Protocol monitoring sites in Saguaro National Park.

IMPROVE thanks Darcy Anderson, the state of Arizona's representative to the IMPROVE Program, who hosted the meeting and coordinated the site tours.

For more information contact Mark Pitchford at NOAA. Telephone: 702/862-5432. Fax: 702/862-5507. E-mail: marcp@snsc.dri.edu.

LADCO releases upper Midwest PM_{2.5} report

The Lake Michigan Air Directors Consortium (LADCO) released a report summarizing current knowledge of ambient PM_{2.5} concentrations in the upper Midwest, or the EPA Region V states (Illinois, Indiana, Michigan, Minnesota, Ohio, and Wisconsin,). Data obtained for the report were collected between 1999 and 2001, using approximately 200 FRM samplers (and other monitors including IMPROVE samplers) in the six-state region. Findings of the study are:

- Non-attainment of the annual average PM_{2.5} concentrations to the National Ambient Air Quality Standards is likely.
- PM_{2.5} concentrations come largely from regional contributions.
- PM_{2.5} concentrations are relatively consistent throughout the year, with some seasonal variation.
- Daily PM_{2.5} concentrations are not above the 24-hour average standard in most areas, but the concentrations do present health problems.
- Sulfates and organics dominate the chemical spectrum.
- Nitrates are important contributors in winter.

The 65-page report was released in June, and can be found on the new LADCO/Midwest Regional Planning Organization Web site, which is currently undergoing redesign, at <http://64.27.125.175>.

For more information contact Mike Koerber at LADCO. Telephone: 847/296-2181. Fax: 847/296-2958. E-mail: koerber@ladco.org.

WESTAR prepares reasonable attribution recommendations for RA-BART

The Western States Air Resources Council (WESTAR) has prepared a report addressing the RA-BART (reasonably attributable visibility impairment and best available retrofit technology) attribution process. The report offers recommendations that may be used to assess reasonable attribution of visibility impairment in Class I areas. These recommendations are:

General principles - Assessments should be:

- A collaborative process that relies on existing data.
- Technically and legally defensible.
- Accomplished at a reasonable cost within reasonable time.
- No more complex than necessary.
- Performed by state or tribal staff.
- Adequate to determine whether or not visibility impairment is attributable to an existing stationary source potentially subject to BART.

Technical criteria - Assessments should:

- Identify the pollutants of concern. Emissions from BART-eligible sources must "cause or contribute to" visibility impairment.
- Consider several factors in assessing impairment, including duration, frequency, geographic extent, magnitude, and time of occurrence.
- Identify distance from the source to the Class I area.
- Use quantitative results (preferably).
- Use as many indicators of impairment as practical.
- Consider the level of uncertainty.
- Use EPA guideline models whenever practical. RA-BART and associated monitoring and modeling techniques.

The report can be found at <http://www.westar.org>. Click on *Files and Downloads*, and look under *Reports* for "Recommendations for Making Determinations in the Context of Reasonably Attributable BART."

For more information contact Bob Lebens at WESTAR. Telephone: 503/387-1660 ext. 6. Fax: 503/387-1671. E-mail: blebens@westar.org.

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IMPROVE STEERING COMMITTEE

IMPROVE Steering Committee members represent their respective agencies and meet periodically to establish and evaluate program goals and actions. IMPROVE-related questions within agencies should be directed to the agency's Steering Committee representative. Steering Committee representatives are:

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* Steering Committee chair

**** Please make a note of Marc Pitchford's new telephone and fax numbers.**

ASSOCIATE MEMBERS

Associate Membership in the IMPROVE Steering Committee is designed to foster additional IMPROVE-comparable visibility monitoring that will aid in understanding Class I area visibility, without upsetting the balance of organizational interests obtained by the steering committee participants. Associate Member representatives are:

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interested in becoming Associate
Members may contact any
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to: assess the impacts of new emission
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progress toward the national visibility
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