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Interpretation of Trends of $PM_{2.5}$ and Reconstructed Visibility from the IMPROVE Network

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ABSTRACT

Under the IMPROVE visibility monitoring network, federal land managers have monitored visibility and fine particle concentrations at 29 Class I area sites (mostly national parks and wilderness areas) and Washington, DC since 1988. This paper evaluates trends in reconstructed visibility and fine particles for the 10th (best visibility days), 50th (average visibility days), and 90th (worst visibility days) percentiles over the nine-year period from 1988–96. Data from these sites provides an indication of regional trends in air quality and visibility resulting from implementation of various emission reduction strategies.

INTRODUCTION

In recent years, the health and environmental effects of particulate matter air pollution have been the subject of much research and discussion by scientific and policy professionals in the air quality community. Significant regulatory programs are under way to address these effects and are expected to result in decreasing ambient particulate matter concentrations (PM) and improved visibility over time.

Implementation of the acid rain program is expected to reduce annual sulfur oxide emissions by more than 10 million tons (from 1980 levels) by 2010.[1] In July 1997, the Environmental Protection Agency (EPA) promulgated new primary and secondary national ambient air quality standards for $PM_{2.5}$ (particulate matter less than 2.5 µm) and EPA proposed a national program to reduce regional haze visibility impairment in more than 150 Class I areas across the country. Implementation of these programs over the coming years will require additional technical assessment and strategy development. Monitoring of particulate matter concentrations, visibility levels, and analysis of trends over time are critical activities needed to develop and evaluate strategies to reduce acid rain, attain national health standards, and make reasonable progress in reducing visibility impairment.

Since 1988, the Interagency Monitoring of Protected Visual Environments (IMPROVE) national visibility monitoring network has been in operation in 29 primarily rural Class I sites and one urban site (Washington, D.C.) across the country (see Figure 1 for site locations). The network is cooperatively managed and funded by the Department of Interior (National Park Service, Fish and Wildlife Service, and Bureau of Land Management), Department of Agriculture (U.S. Forest Service), the Environmental Protection Agency, and state governments. This paper uses data collected in the IMPROVE monitoring program to examine nine-year trends (1988–1996) in Class I area visibility and $PM_{2.5}$ concentrations.

> **IMPLICATIONS**
> The visibility protection program under the Clean Air Act as amended in 1990 is guided by a national goal that calls for “the prevention of any future, and the remedying of any existing, [anthropogenic] impairment of visibility in mandatory Class I Federal areas.” Trends over time in monitored visibility and fine particulate concentrations will help assess whether emission reduction strategies are achieving their desired effect.

AEROSOL MONITORING

The IMPROVE aerosol sampler was designed specifically for the IMPROVE monitoring program. Analytical methods and filter media used to determine the chemical composition of the particles are discussed by Eldred et al.[2] and summarized in Table 1. Two 24-hour samples are collected each week, on Wednesday and Saturday, beginning midnight local time. The fine aerosol species found at most continental sites can be classified into five major chemical types: sulfates, nitrates, mass associated with organic carbon (OMC), light-absorbing carbon (LAC), and soil.[3,4] The fine aerosol types are composites of the elements and

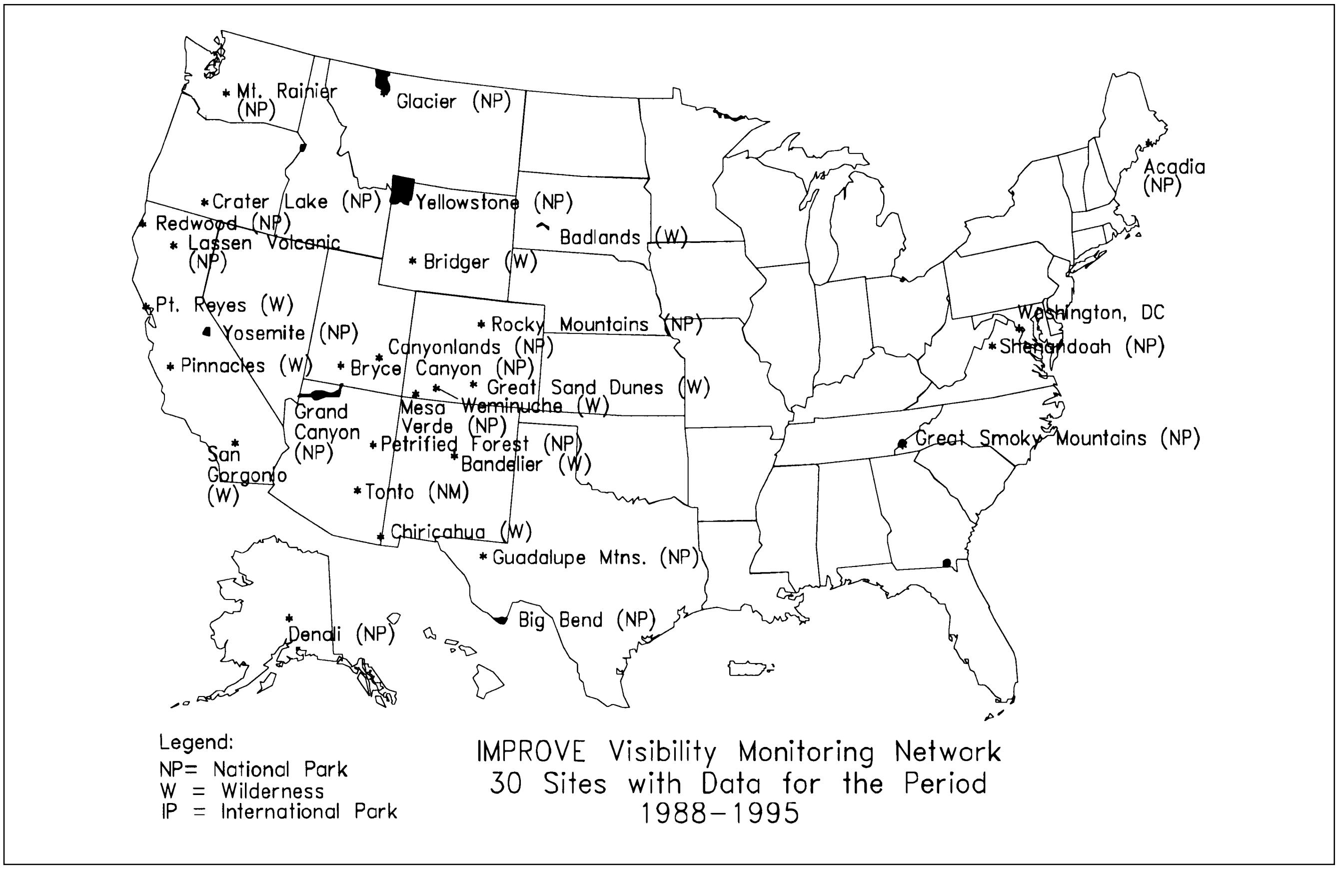

Figure 1. The 30 IMPROVE monitoring sites in operation from 1988 to 1995 equipped with Channels A–D.

ions measured in IMPROVE samplers, and their concentrations or masses are estimated from the masses of the measured elements and ions according to their presumed or probable composition as summarized by Table 2.

RECONSTRUCTED LIGHT EXTINCTION COEFFICIENT

The extinction coefficient, b_{ext} (expressed as inverse megameters, 1/Mm), is the sum

$$b_{ext} = b_{sp} + b_{ap} + b_{sg} + b_{ag} \quad (1)$$

where light scattering by gases in the atmosphere, b_{sg}, is described by the Rayleigh scattering theory[5] and will be referred to as Rayleigh scattering, the IMPROVE program assumes a standard value of 10/Mm; absorption due to gases, b_{ag}, is primarily due to NO_2 and is assumed to be negligible because almost all monitoring sites are in rural locations;[6] absorption by particles, b_{ap}, is caused primarily by carbon containing particles, b_{sp}, which is caused by both fine and coarse aerosol species and is the largest contributor to total light extinction in most locations.[7]

A particle in the atmosphere can be a mix (internal mixture) of various aerosol species or in some cases its compositional structure may be restricted to one species (external mixture) such as $(NH_4)_2SO_4$. Furthermore, an internally mixed aerosol such as organic/sulfate/water particle can be externally mixed from wind blown dust particles. Whether a particle is internally or externally mixed it scatters and/or absorbs a specific fraction of radiant energy impinging on it. Following the suggestion of White[8] its scattering/extinction per unit mass ratio will be referred to as specific scattering/extinction, as in specific gravity.

Most routine aerosol monitoring programs and many special study visibility characterization programs were

Table 1. Analysis techniques used to determine chemical composition of particulate matter in the IMPROVE network.

Module	Filter	Analysis
A	Teflon	Gravimetric analysis for $PM_{2.5}$, LIPM for optical absorption (b_{abs}), PIXE for elements Na to Pb, PESA for H.
B	nylon (denuded)	Ion chromatography for NO_3.
C	Quartz	TOR for organic and elemental C.
D	Teflon	Gravimetric analysis for PM_{10}.

LIPM = Laser Integrating Plate Method for b_{abs}; PESA = Proton Elastic Scattering; PIXE = Particle Induced X-ray Emission; TOR = Thermal Optical Reflectance.

Table 2. The formulae and assumptions applied to IMPROVE sampler measurements to derive the principal fine aerosol species, reconstructed fine mass, and coarse mass. The brackets indicate the mass concentration of the aerosol species or element.

Species	Formula	Assumptions
SULFATE	4.125[S]	All elemental S is from sulfate. All sulfate is from ammonium sulfate.
NITRATE	1.29[NO_3]	Denuder efficiency is close to 100%. All nitrate is from ammonium nitrate.
LAC (Light-absorbing carbon by channel C)	[ECLT] + [ECHT]	All high temp carbon is elemental
OMC (Organic mass from carbon)	1.4{[OCLT]+[OCHT]}	Average organic molecule is 70% carbon.
SOIL (Fine soil)	2.2[Al]+2.19[Si] +1.63[Ca]+2.42[Fe] +1.94[Ti]	[Soil K]=0.6[Fe]. FeO and Fe_2O_3 are equally abundant. A factor of 1.16 is used for MgO, Na_2O, H_2O, CO_2.
RCFM (Reconstructed fine mass)	[SULFATE]+[NITRATE] +[LAC]+[OMC]+[SOIL]	Represents dry ambient fine aerosol mass for continental sites.
CM (Coarse mass)	[PM_{10}] - [$PM_{2.5}$]	Consists only of insoluble soil particles.

designed to measure bulk aerosol species mass concentrations such as sulfates, nitrates, carbonaceous material, and selected elements.[9-13] They were not designed to determine the micro-physical and chemical characteristics of these species.

The inherent limitations of estimating aerosol optical properties from bulk aerosol measurements have been addressed, at least in part, by a number of authors. For instance, Ouimette and Flagan[14] have shown, from basic theoretical considerations, that if an aerosol is mixed externally or if in an internally mixed aerosol the index of refraction is not a function of composition or size, and the aerosol density is independent of volume, then

$$b_{ext} = \sum_i a_i m_i \qquad (2)$$

where a_i is the specific scattering or absorption efficiency and m_i is the mass of the individual species.

Malm and Kreidenweis[15] demonstrated from a theoretical perspective, that specific scattering of mixtures of organics and sulfates were insensitive to the choice of internal or external mixtures. Sloane[16-18] and Sloane and Wolff,[19] and more recently Lowenthal et al.,[20] Malm et al.,[21] and Malm et al.[22] have shown that differences in estimated specific scattering between external and internal model assumptions are usually less than about 10%. In the absence of detailed microphysical and chemical structure of ambient aerosols the above studies demonstrate that a reasonable estimate of aerosol scattering can be achieved by assuming that each species is externally mixed.

However, the issue of water uptake by hygroscopic species must be addressed. Implicit to the use of eq 2 is an assumed linear relationship between aerosol mass and extinction. It is well known that sulfates and other hygroscopic species uptake water and grow nonlinearly as relative humidity increases and therefore if scattering is measured at various relative humidities the relationship between measured scattering and hygroscopic species mass can be quite nonlinear. A number of authors have attempted to linearize the model, in an empirical way, by multiplying the hygroscopic species by such a factor as 1/(1-RH) to account for the presence of water mass.[23,24] However, Malm et al.[25] and Gebhart and Malm[26] proposed a different approach. They multiplied the hygroscopic species by a relative humidity scattering enhancement factor, f(RH), that is calculated on a sampling-period-by-sampling-period basis using Mie theory and an assumed size distribution and laboratory measured D/D_o curves.

Because mixtures of ammoniated sulfate compounds with other species have been shown to be hygroscopic below the deliquescent values[17,18,27] and because the growth factor and light-scattering efficiency for ambient aerosols has previously been observed to be rather smooth,[16-18,28-30] a "best estimate" for the sulfate and nitrate species growth, the D/D_o laboratory growth curves, as measured by Tang[31] that demonstrate hysteresis, were smoothed between the deliquescence and crystallization points. Malm et al.[21,22] have demonstrated that in both the East (Great Smoky Mountains National Park) and West (Grand Canyon National Park) the best estimate growth model used to develop a theoretically f(RH) function yields good agreement between measured and reconstructed scattering for particles less than 2.5 μm.

Therefore, the following equation is used to estimate reconstructed particle scattering:

$$\begin{aligned} b_{scat} &= (3)f(RH)[SULFATE] \\ &+ (3)f(RH)[NITRATE] \\ &+ (4) f_{org}(RH)[OMC] \\ &+ (1)[SOIL] + (0.6)[CM] \end{aligned} \qquad (3)$$

The brackets indicate the species concentration, 3 m^2/g is the dry specific scattering for sulfates, and nitrates, 4 m^2/g for organic carbon, and 1 m^2/g and 0.6 m^2/g are the respective scattering efficiencies for soil and coarse mass. The efficiencies for fine soil and coarse mass are taken from a literature review by Trijonis and Pitchford.[6]

A dry scattering efficiency of 3 m^2/g is a nominal scattering efficiency based on a literature review by Trijonis et al.[32,33] and a review by White.[34] Trijonis' best estimate for sulfates and nitrates is 2.5 m^2/g with an error factor of 2, while for organics it is 3.75 m^2/g again with an error factor of 2. White took a somewhat different approach in that he reviewed 30 studies in which particle scattering and mass were measured. He then estimated a high and low scattering efficiency by using mass measurements to prorate the measured extinction. For sulfate the low estimate was arrived at by assuming that sulfate, nitrate, and organics scatter twice as efficiently as all other species, and for the high estimate he assumed that only sulfate was twice as efficient. His low and high sulfate mass scattering efficiencies for the rural west were 3.0 and 3.7 m^2/g, respectively. For organics his low estimate assumes that organics and other non-sulfate species scatter half as efficiently as sulfates, and for the high estimate he assumes organics are three, and sulfates twice as efficient at scattering light as other species. His low and high estimates for organic mass scattering coefficients are 1.8 and 4.1 m^2/g. More recently Malm et al.[35] demonstrated that an assumption dry specific scattering values given in eq 3 yielded good agreement between measured and reconstructed extinction across the whole IMPROVE monitoring network.

Various functions for the hygroscopicity of organics have been proposed. Assumptions must not only be made about the solubility of organics but also on the fraction of organics that are soluble. It should be noted that models that treat water uptake for nonideal, multicomponent solutions using theoretical and semi-theoretical thermodynamic relationships have been developed and have been applied to both visibility and climate forcing problems.[36-39] The correct treatment of the hygroscopicity of species in multicomponent mixtures—especially organic species—remains problematic, not only because of the lack of suitable mixture thermodynamic data, but also because of the lack of information about other critical mixture properties. Given the variety of organic species, it is possible that a geographic variation in organic species exists, with large fractions of soluble species occurring in certain parts of the continent and much smaller fractions in other areas. However, field experiments and subsequent data analysis at Great Smoky and Grand Canyon National Parks[22,40] and more generally data collected in the IMPROVE network[35] show that to within the uncertainty of the measurements and modeling assumptions organics are not or only weakly hygroscopic. Therefore $f_{org}(RH)$ for organics was set equal to one.

Equation 3 has been shown to give a good estimation of scattering for particles less than 2.5 µm, however, estimating extinction requires knowledge of particle absorption. Mass absorption efficiencies of carbon vary by more than a factor of two as do direct measurements. Horvath[41] has reviewed the measurement of absorption, while Fuller[42] has theoretically explored the variability of absorption efficiency as a function of carbon morphology. Although absorption can be estimated in a variety of ways, there is no one method that is generally accepted by the scientific community. For purposes of this paper carbon absorption is estimated using:

$$b_{abs} = 10LAC \quad (4)$$

where b_{abs} is particle absorption and LAC is the concentration of light-absorbing carbon as measured using the TOR analysis scheme.[43]

Because aerosol concentrations are derived from averages over long periods, the light extinction due to soluble species is derived using hourly RH values less than or equal to 98%, as given by the following equation:

$$b_{scat} = \beta F_T \bar{C} \quad (5)$$

where $\bar{C}$ is the average species concentration, β is the specific scattering, and

$$F_T = \overline{f_T(RH)} \quad (6)$$

Using eq 3, extinction budgets for a time interval may be calculated by replacing f(RH) with F_T and by using the average concentration of each species over the same time interval as the mass concentration.

Using the data from sites with collocated optical and RH data, a polynomial curve was fitted to the annual and seasonal data as defined by

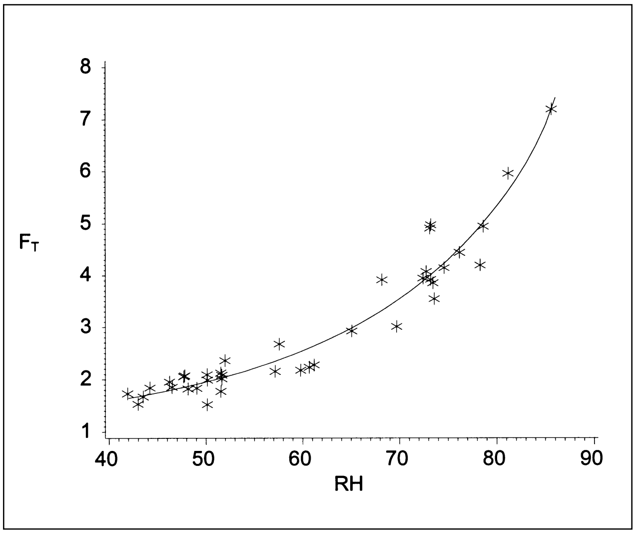

Figure 2. Relationship between average site relative humidity (RH) and the relative humidity correction factor (F_T) for the 39 IMPROVE sites having co-located relative humidity measurements.

$$F = b_0 + b_2(100/(100 - \overline{RH}))^2 + b_3(100/(100 - \overline{RH}))^3 + b_4(100/(100 - \overline{RH}))^4, \quad (7)$$

where b_o = 0.51757, b_2 = 0.52588, b_3 = -0.09467, and b_4 = -0.005996 with an R-square of 0.94. Figure 2 shows the fitted curve plotted against annual average RH for IMPROVE sites with collocated RH data. For those sites without collocated optical and RH data the annual factors can be calculated using eq 7 and estimates of annual average RH. In this fashion, all 30 sites are treated the same enabling the largest spatial coverage.

The deciview (dv) is a visibility metric based on the light extinction coefficient that expresses incremental changes in perceived visibility.[44] Because the deciview expresses a relationship between changes in light extinction and perceived visibility, it can be useful in describing visibility trends. A one dv change is about a 10% change in extinction coefficient, which is a small but perceptible scenic change under many circumstances. The deciview is defined by the following equation:

$$dv = 10\ln(b_{ext}/10) \quad (8)$$

The deciview scale is near zero for pristine atmosphere (dv = 0 for Rayleigh condition at about 1.8 km elevation) and increases as visibility is degraded.

TRENDS ANALYSIS

Overall Regional Conditions

Because of the significant regional variation in visibility conditions, this section groups the 30 sites with data from 1988 to 1996 into eastern and western "regions." In the east, outside of Washington, DC, two regions are presented: the Northeast, represented by Acadia National Park, and Appalachia, consisting of Shenandoah and Great Smoky Mountains National Parks. In the west, there are the Northern Great Plains, West Texas, Sonora, Colorado Plateau, Central Rockies, Northern Rockies, Cascade, Sierra-Humbolt, West Coast, Sierra, Southern California and Alaska. Table 3 lists the regions and sites assigned to each.

Figures 3a and 3b show the average fine particle and visibility conditions across the United States as pie charts where the pies are scaled to $PM_{2.5}$ and reconstructed extinction, respectively. Table 4 presents the mass and extinction values associated with each species for every region. The contrast between eastern and western conditions is quite evident. There is a factor of three difference in $PM_{2.5}$ loadings between the Colorado Plateau and Applachia at 3.15 $\mu g/m^3$ and 10.81 $\mu g/m^3$, respectively. Alaska is the region with the least amount of $PM_{2.5}$ mass with an annual average of 1.71 $\mu g/m^3$. The eastern regions are dominated by sulfate mass, while in the western sites elemental carbon and organics taken together frequently contribute the most to fine mass concentrations. Southern California is unique, as it is the only region dominated by ammonium nitrate.

For extinction, the east-west differences are even greater, due to the effect of regional variations in relative humidity. The average relative humidity in the east in many regions exceeds 70%, while the western United States has many regions that are very arid. However, it should be noted that Mount Rainier National Park has the highest average humidity of the network. Relative humidity increases the contribution from the soluble nitrates and sulfates to extinction. In the Appalachian region, the fine mass loading is about 60% sulfate but because of relatively high RH conditions the contribution to extinction is much higher at 74% in spite of the fact that reconstructed extinction includes absorption and coarse mass scattering, while fine mass loading calculations do not. There is more than a factor of 5 difference between the Colorado Plateau with an average extinction of 17.3/Mm and Appalachia with an annual average extinction of 97.5/Mm. While the effect of RH is less in the west, it is sufficient to put sulfate on a par with elemental carbon and organic species taken together as the dominant contributor to extinction. As with the case for mass loading southern California is unique in that ammonium nitrate is the dominant contributor to extinction.

Trends over Time

Stimulated by rollbacks in SO_2 emissions many authors have examined trends in air quality.[45-47] Hussain et al.[45] looked at wet deposition data from Whiteface Mountain and Mayville in New York State and found average sulfate ion concentrations have decreased by 47% and 30%, respectively, between 1979 to 1996. Lynch et al.[46,47] have examined trends in wet deposition ion concentrations using data from the NADP National Trends Network sites east of the Mississippi River and have found widespread declines of sulfate ion concentrations and significant decreases in base cation concentration.

Only recently has the IMPROVE aerosol network, initiated in March 1988, matured to a point where long-term trends of average ambient aerosol concentrations and reconstructed extinction can be assessed. Also, changes over time of the distribution of aerosol concentrations and extinction can be studied leading to an understanding of trends in clear and hazy days in addition to median or average conditions.

Characterization of trends, however, with only nine years of data can be a highly subjective exercise in that slopes and their significance can vary depending on the technique employed. Using the ordinary least-squares (OLS) regression approach is questionable with such small data sets as the results can be highly influenced by outliers

Table 3. Regions and IMPROVE monitoring sites with data starting in March 1988 assigned to each region.

Region	Sites
Alaska	Denali National Park
Appalachia	Shenandoah National Park, Great Smoky Mountains National Park
Cascades	Mount Rainier National Park
Colorado Plateau	Petrified Forest National Park; Hopi Point, AZ; Bryce Canyon National Park
	Mesa Verde National Park, Bandelier National Monument, Canyonlands National Park
Central Rockies	Yellowstone National Park, Bridger Wilderness, Rocky Mountain National Park
	Great Sand Dunes National Monument, Weminuche Wilderness
Coastal	Pinnacles National Monument, Point Reyes National Seashore
Northeast	Acadia National Park
Northern Great Plains	Badlands National Park
Northern Rockies	Glacier National Park
Southern California	San Gorgonio Wilderness
Sonora	Chiricahua National Monument, Tonto National Monument
Sierra Nevada	Yosemite National Park
Sierra Humbolt	Lassen Volcanic National Park, Crater Lake National Park
Washington DC	Washington, DC
West Texas	Guadalupe Mountains National Park, Big Bend National Park

plus standard deviations and standard errors can be large. In another approach, developed by Theil,[48] outlier data points do not influence the results. Slopes of trend lines are calculated for each site by first finding the slope between all possible pairs of data points, then sorting the results from smallest or most negative to the largest and finally, the median value in the case of an odd number of pairs is selected as the estimated slope, or in the case of even number of pairs the average of the two slopes that straddle the median is used as the estimate. The significance of the Theil slope is found by assuming that the "true" slope is zero, then calculating the probability that the estimated slope occurred by chance. This technique has also been adopted by the Environmental Protection Agency (EPA) for estimating trends in air quality data.[49]

Iyer et al.[50] recently used data collected in the IMPROVE network to study the evolution over time of the distribution of aerosol concentrations finding that the frequency of occurrence of days with high concentrations of $PM_{2.5}$, elemental carbon, organics, and sulfate has decreased at many sites accompanied by an increase in the frequency of low concentration days.

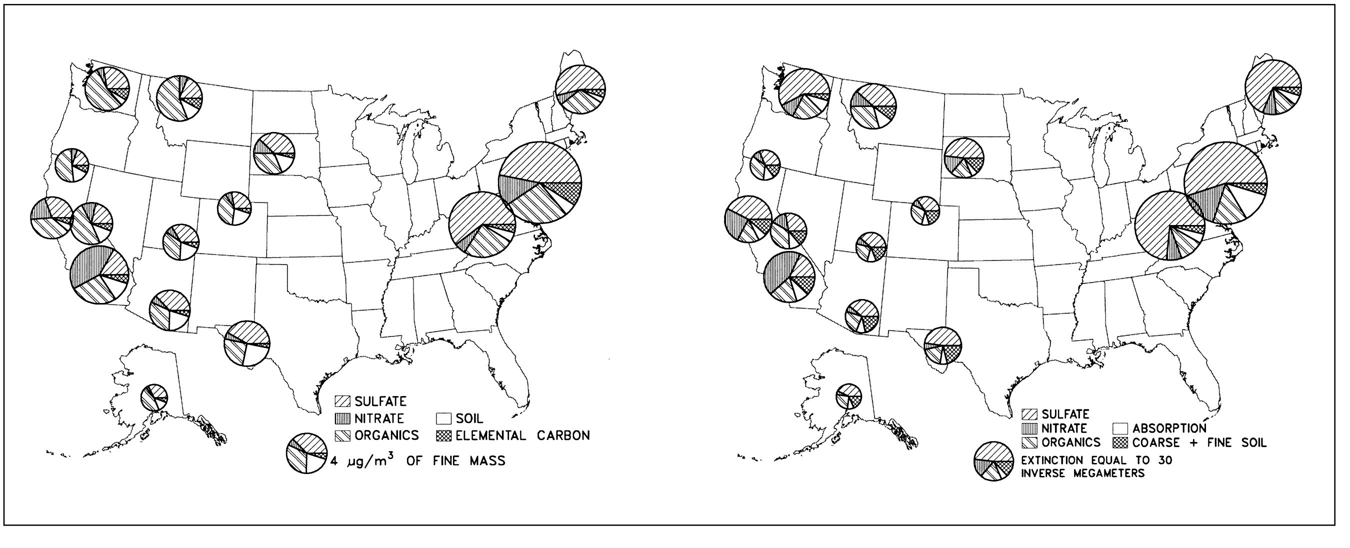

Figure 3. Pie charts showing the regional distribution of aerosol $PM_{2.5}$ and extinction conditions and contributions from principal aerosol species. $PM_{2.5}$ is due to sulfate, nitrate, organics, fine soil, and elemental carbon. Aerosol extinction is contributed to by sulfate extinction, nitrate extinction, extinction from organic species, absorption, and extinction from coarse mass plus fine soil.

Table 4. Regional $PM_{2.5}$ and extinction summaries. $PM_{2.5}$ has been broken down into the principal subspecies: sulfate, nitrate, organic mass, fine soil, and elemental carbon. Aerosol extinction has been broken down to principal contributing species: sulfate extinction, nitrate extinction, organic mass extinction, absorption, and extinction from coarse mass plus fine soil.

Annual $PM_{2.5}$ Region	$PM_{2.5}$	Sulfate	Nitrate	Organics	Fine Soil	Elemental Carbon
Alaska	1.71	0.55	0.06	0.77	0.22	0.10
Appalachia	10.81	6.53	0.60	2.73	0.52	0.43
Cascades	4.67	1.30	0.23	2.51	0.22	0.41
Colorado Plateau	3.15	1.06	0.21	1.08	0.64	0.17
Central Rockies	2.87	0.80	0.18	1.11	0.64	0.14
Coastal	4.40	1.35	0.90	1.65	0.25	0.25
Northeast	6.13	3.32	0.40	1.84	0.23	0.34
Northern Great Plains	4.26	1.61	0.51	1.35	0.63	0.16
Northern Rockies	5.15	0.98	0.31	2.88	0.57	0.41
Southern California	8.64	1.45	3.53	2.29	0.94	0.42
Sonora	4.09	1.52	0.24	1.28	0.84	0.20
Sierra Nevada	4.40	0.96	0.47	2.16	0.55	0.26
Sierra Humboldt	2.67	0.52	0.16	1.36	0.42	0.20
Washington, DC	16.90	7.91	2.16	4.44	0.82	1.56
West Texas	5.11	2.13	0.25	1.29	1.27	0.17

Annual Extinction Region	Aerosol Extinction	Sulfate	Nitrate	Organic	Absorption	Coarse + Fine Soil
Alaska	11.9	5.1	0.6	3.1	1.0	2.2
Appalachia	97.6	71.7	6.9	10.9	4.3	3.8
Cascades	50.6	29.1	5.0	10.0	4.1	2.3
Colorado Plateau	17.3	6.7	1.3	4.3	1.7	3.3
Central Rockies	15.8	5.5	1.2	4.4	1.4	3.2
Coastal	43.5	18.4	10.9	6.6	2.5	5.1
Northeast	59.3	40.6	4.8	7.3	3.4	3.0
Northern Great Plains	30.3	14.6	4.7	5.4	1.6	4.0
Northern Rockies	39.5	15.0	4.7	11.5	4.1	4.1
Southern California	51.7	9.3	22.6	9.2	4.2	6.3
Sonora	21.3	8.3	1.3	5.1	2.0	4.6
Sierra Nevada	25.2	7.0	3.5	8.6	2.6	3.5
Sierra Humboldt	16.7	5.2	1.5	5.5	2.0	2.5
Washington, DC	132.8	73.2	19.9	17.8	15.6	6.3
West Texas	27.0	12.9	1.5	5.2	1.7	5.7

In this paper, we examine trends of the distribution of $PM_{2.5}$ mass concentrations and reconstructed extinction expressed as deciview using the technique suggested by Theil. The trends are examined by sorting each year's data into three groups based on the cumulative frequency of occurrence of $PM_{2.5}$: best visibility days, 0–20%; median, 40–60%; and worst visibility days, 80–100%. Each group was then labeled by its mid point (e.g., 10th, 50th, 90th percentiles). After sorting each group's average concentrations of $PM_{2.5}$ and identifying the associated principal aerosol species, reconstructed light extinction and deciview can be calculated

In addition to statistical concerns, there is always the underlying year-to-year variability due to meteorology. While the importance of meteorologically induced trends cannot be ignored, when trying to deduce the effect of emission rollbacks, the roll of meteorology is beyond the scope of this article.

Results of the Theil slope estimates for the 90, 50, and 10 subgroups are presented in Tables 5, 6, and 7, respectively. Estimates are given for both deciview and $PM_{2.5}$ and each slope is paired with the probability for rejection.

Selected Examples of Trends in $PM_{2.5}$ and Deciview. Figure 4 shows plots of the 10, 50, and 90 groups at Pinnacles National Monument for both $PM_{2.5}$ and deciview. The annual data has SYEAR (sample year beginning in March of each year) on the horizontal axis. Pinnacles is the only site that has significant negative slopes for all subgroups for both deciview and $PM_{2.5}$. All annual slopes were significant at the 0.05 level or better.

Figure 5 shows the same plots for Badlands National Park, the only site with a significant group 90 positive slope at the 0.1 level for deciview. This site contrasted with Pinnacles demonstrates the difficulty associated with determining trends with only nine years of data. Although the group 90 slope was positive and significant at the 0.1 level, one might draw a different conclusion by focusing only on the years 1992 through 1996. It is also of interest to note that although $PM_{2.5}$ in 1996 is less than in 1988 calculated deciview is higher in 1996 than in 1988; in fact, the annual slope for deciview is positive at 0.065 dv/yr but the annual slope for $PM_{2.5}$ is negative at -0.033 mg/m^3 although insignificant. Since the same correction of RH was used for all the years this suggests that the aerosol mix has changed over time with increasing concentrations of soluble species or light-absorbing carbon. For the 50 and 90 groups, all coefficients are uniformly negative and mostly significant.

Figure 6 shows the data for Big Bend National Park, which had insignificant trends in deciview for all three subgroups with group 90 being positive and the others negative. $PM_{2.5}$ shows a similar pattern with slightly more significance with an annual slope of –0.0526 mg/m^3 and significant at the 0.1 level. The group 90 data is "U" shaped, with the bottom occurring in 1992, reinforcing the difficulty of assessing long-term trends with so few data points. One would obviously draw different conclusions by focusing on the years 1988 through 1992 versus the years 1992 through 1996.

Trends in $PM_{2.5}$ and Deciview Across the United States. U.S. maps summarizing results of the trend analysis for the annual deciview data are shown in Figures 7, 8, and 9 for

Table 5. Worst 20%–Group 90. Annual slope estimates with probabilities for rejection using Theil's method applied to deciview and $PM_{2.5}$ for the average of the worst 20% days.

Site	Deciview Annual Slope	Deciview Annual Prob	$PM_{2.5}$ Annual Slope	$PM_{2.5}$ Annual Prob
Acadia NP	-0.065	0.238	-0.185	0.060
Badlands NP	0.069	0.060	-0.033	0.179
Bandelier NM	-0.027	0.381	-0.024	0.306
Big Bend NP	0.066	0.130	0.074	0.130
Bryce Canyon NP	-0.017	0.381	-0.023	0.238
Bridger W	-0.124	0.306	-0.028	0.460
Canyonlands NP	-0.162	0.060	-0.117	0.060
Chiricahua NM	0.000	0.540	0.004	0.540
Crater Lake NP	0.073	0.306	-0.005	0.460
Denali NP	-0.138	0.090	-0.091	0.179
Glacier NP	-0.093	0.090	-0.112	0.060
Grand Canyon NP	-0.072	0.238	-0.103	0.179
Great Sand Dunes NM	-0.183	0.179	-0.060	0.381
Great Smoky Mountains NP	0.086	0.179	0.169	0.130
Guadalupe Mountains NP	-0.088	0.130	0.066	0.238
Lassen Volcanic NP	-0.039	0.381	-0.028	0.460
Mesa Verde NP	0.000	0.540	-0.009	0.460
Mount Rainier NP	-0.162	0.060	-0.197	0.022
Petrified Forest NP	-0.146	0.060	-0.112	0.022
Pinnacles NM	-0.208	0.006	-0.210	0.003
Point Reyes NS	-0.224	0.090	-0.265	0.038
Redwood NP	-0.131	0.060	-0.206	0.001
Rocky Mountain NP	-0.033	0.306	-0.003	0.540
San Gorgonio W	-0.194	0.022	-0.374	0.012
Shenandoah NP	0.006	0.540	-0.085	0.381
Tonto NM	-0.040	0.306	-0.040	0.381
Washington	-0.014	0.460	0.075	0.381
Weminuche W	-0.133	0.022	-0.106	0.130
Yellowstone NP	-0.171	0.306	-0.038	0.381
Yosemite NP	0.066	0.381	0.122	0.179

NP = National Park; NM = National Monument; W = Wilderness Area; NS = National Seashore.

Table 6. Median 20%–Group 50. Annual slope estimates with probabilities for rejection using Theil's method applied to deciview and $PM_{2.5}$ for the average of the median 20% days.

Site	Deciview Annual Slope	Deciview Annual Prob	$PM_{2.5}$ Annual Slope	$PM_{2.5}$ Annual Prob
Acadia NP	-0.150	0.022	-0.103	0.006
Badlands NP	-0.113	0.012	-0.123	0.003
Bandelier NM	-0.178	0.003	-0.087	0.001
Big Bend NP	-0.020	0.179	-0.053	0.090
Bryce Canyon NP	-0.107	0.090	-0.064	0.022
Bridger NM	-0.091	0.130	-0.061	0.012
Canyonlands NP	-0.116	0.038	-0.093	0.022
Chiricahua NM	-0.061	0.060	-0.053	0.060
Crater Lake NP	-0.042	0.460	-0.064	0.130
Denali NP	-0.157	0.090	-0.052	0.022
Glacier NP	0.123	0.022	-0.135	0.000
Grand Canyon NP	-0.114	0.130	-0.089	0.006
Great Sand Dunes NM	-0.200	0.022	-0.096	0.038
Great Smoky Mountains NP	-0.064	0.381	-0.110	0.238
Guadalupe Mountains NP	-0.057	0.238	-0.057	0.090
Lassen Volcanic NP	-0.147	0.006	-0.077	0.012
Mesa Verde NP	-0.056	0.306	-0.025	0.179
Mount Rainier NP	-0.029	0.460	-0.090	0.090
Petrified Forest NP	-0.183	0.022	-0.106	0.012
Pinnacles NM	-0.206	0.003	-0.155	0.012
Point Reyes NS	-0.208	0.022	-0.127	0.022
Redwood NP	-0.131	0.012	-0.087	0.001
Rocky Mountain NP	-0.174	0.012	-0.085	0.001
San Gorgonio W	-0.139	0.130	-0.235	0.130
Shenandoah NP	-0.129	0.090	-0.181	0.003
Tonto NM	-0.080	0.060	-0.070	0.022
Washington	-0.035	0.460	-0.087	0.460
Weminuche W	-0.118	0.012	-0.069	0.001
Yellowstone NP	-0.121	0.130	-0.070	0.038
Yosemite NP	-0.073	0.238	-0.041	0.238

NP = National Park; NM = National Monument; W = Wilderness Area; NS = National Seashore.

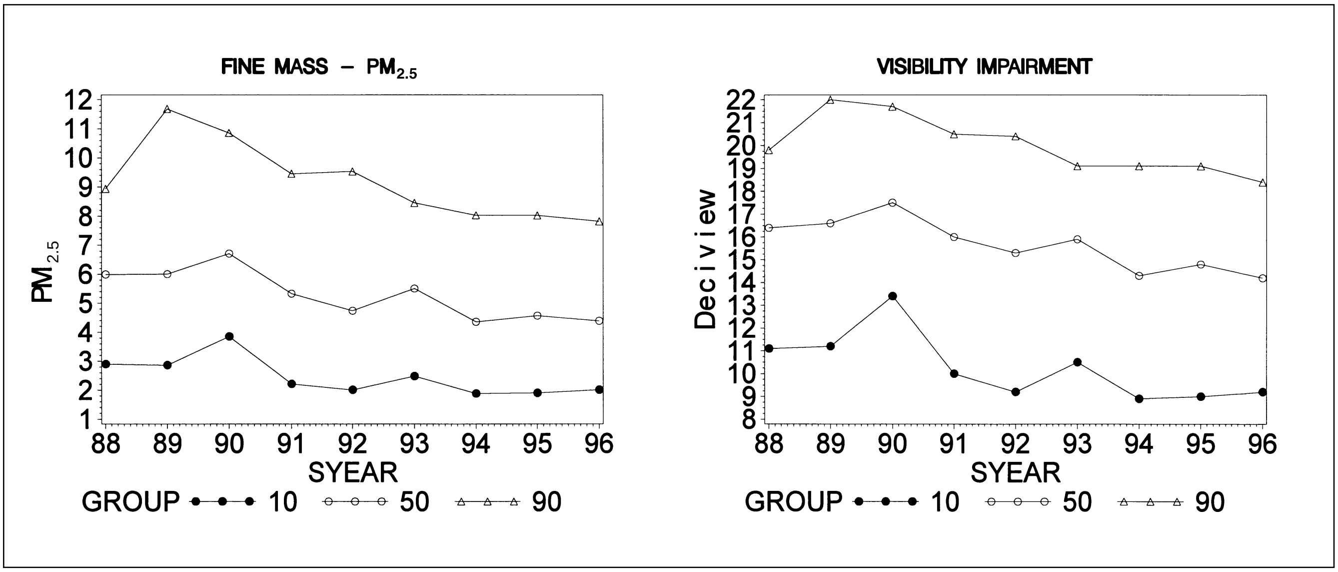

Figure 4. Plots of the 10, 50, and 90 groups for both $PM_{2.5}$ and deciview at Pinnacles National Monument.

Table 7. Best 20%–Group 10. Annual slope estimates with probabilities for rejection using Theil's method applied to deciview and $PM_{2.5}$ for the average of the best 20% days.

Site	Deciview Annual Slope	Deciview Annual Prob	$PM_{2.5}$ Annual Slope	$PM_{2.5}$ Annual Prob
Acadia NP	-0.244	0.012	-0.104	0.006
Badlands NP	-0.100	0.130	-0.073	0.001
Bandelier NM	-0.171	0.060	-0.061	0.012
Big Bend NP	-0.085	0.130	-0.061	0.238
Bryce Canyon NP	-0.134	0.012	-0.058	0.001
Bridger	-0.106	0.090	-0.031	0.038
Canyonlands NP	-0.151	0.012	-0.083	0.001
Chiricahua NM	-0.007	0.460	-0.030	0.179
Crater Lake NP	-0.167	0.012	-0.061	0.001
Denali NM	-0.164	0.038	-0.061	0.003
Glacier NP	-0.106	0.238	-0.069	0.001
Grand Canyon NP	-0.100	0.060	-0.058	0.003
Great Sand Dunes NM	-0.269	0.006	-0.091	0.006
Great Smoky Mountains NP	-0.062	0.306	-0.082	0.060
Guadalupe Mountains NP	-0.075	0.238	-0.048	0.179
Lassen Volcanic NP	-0.168	0.022	-0.056	0.006
Mesa Verde NP	-0.146	0.060	-0.035	0.012
Mount Rainier NP	-0.183	0.130	-0.056	0.022
Petrified Forest NP	-0.176	0.012	-0.084	0.006
Pinnacles NM	-0.207	0.038	-0.083	0.022
Point Reyes NS	-0.074	0.179	-0.034	0.060
Redwood	-0.194	0.012	-0.057	0.006
Rocky Mountain NP	-0.139	0.038	-0.063	0.003
San Gorgonio W	-0.030	0.460	-0.038	0.381
Shenandoah NP	-0.001	0.540	-0.040	0.179
Tonto NP	-0.139	0.022	-0.062	0.022
Washington	-0.129	0.238	-0.101	0.238
Weminuche W	-0.008	0.460	-0.038	0.006
Yellowstone NP	-0.325	0.003	-0.105	0.001
Yosemite NP	-0.061	0.130	-0.039	0.060

NP = National Park; NM = National Monument; W = Wilderness Area; NS = National Seashore.

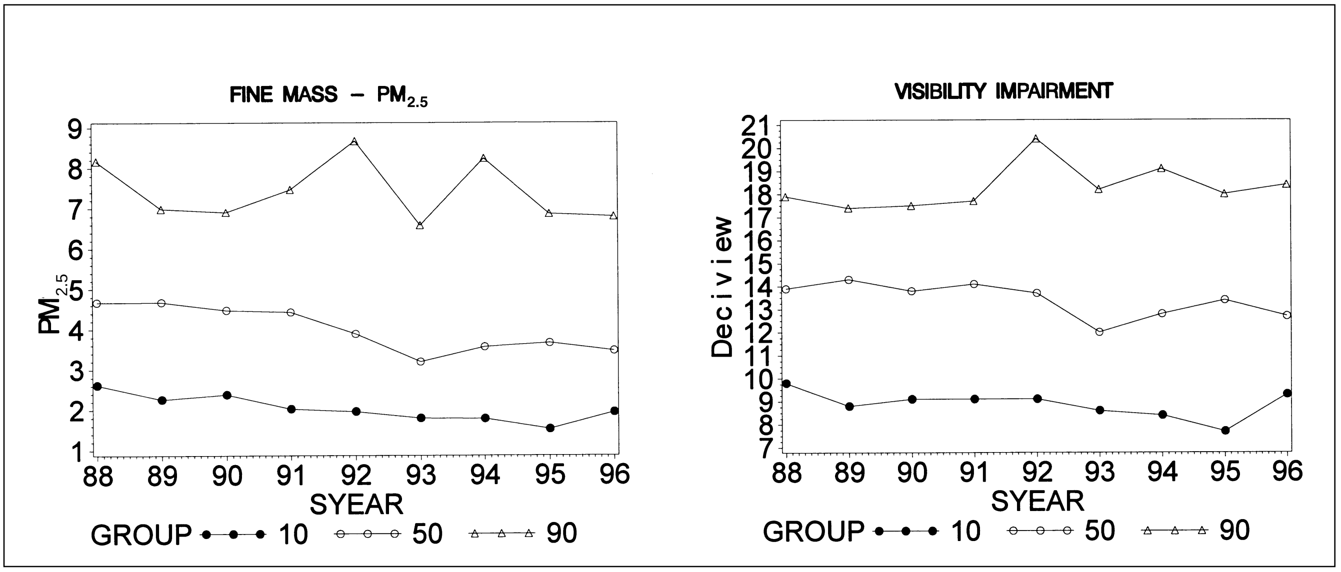

Figure 5. Plots of the 10, 50, and 90 groups for both $PM_{2.5}$ and deciview at Badlands National Park.

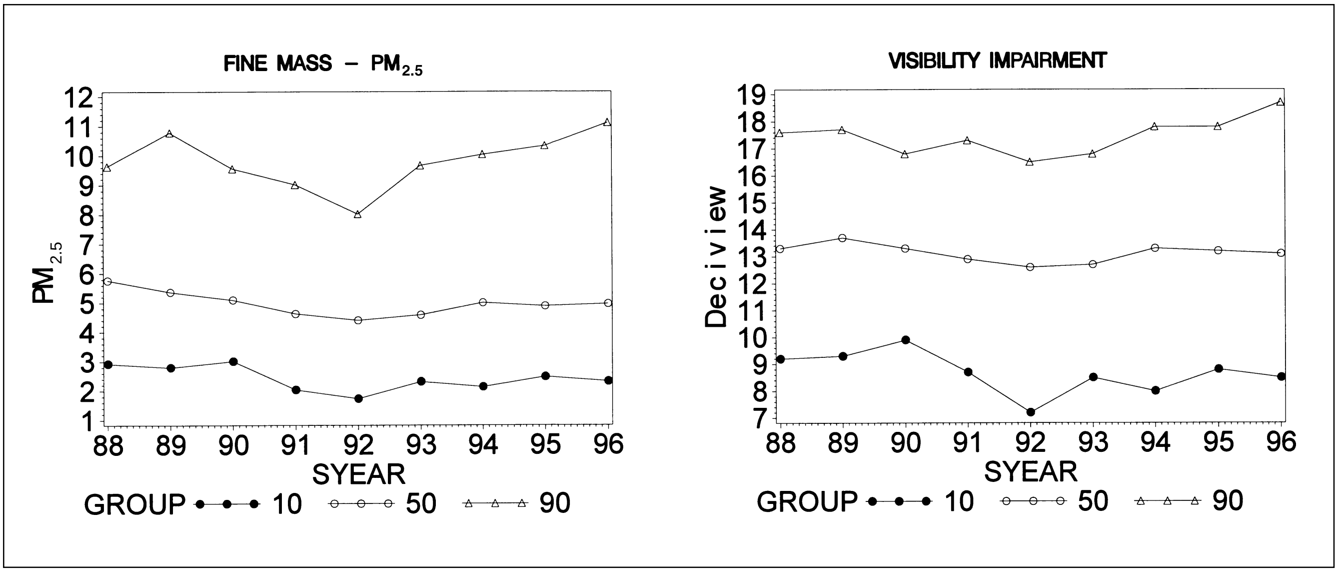

Figure 6. Plots of the 10, 50, and 90 groups for both $PM_{2.5}$ and deciview at Big Bend National Park.

the 90, 50, and 10 subgroups, respectively. The icons mark the site locations, a star indicates an insignificant slope, the empty triangles indicate a positive or negative slope significant at the 0.1 level of probability, and the solid triangle is significant at the 0.05 level. A decreasing trend indicates, in deciview, improving visibility, while an increasing trend indicates worsening visibility.

Group 90 (Figure 7, Table 5), the worst days, has eight sites with positive slopes with all but one having insignificant slopes. The site at Badlands, representing the Northern Great Plains region, has a slope of 0.069 dv/yr at the 0.1 level of significance. In the east, Washington, DC and Acadia and in the west, Big Bend, Mesa Verde, Yosemite, and Crater Lake National Parks and Chiricahua National Monument, show positive but insignificant slopes.

At the remaining 20 western sites, all slopes are negative with a subset of nine being significant. California boasts the most sites showing significance, with San Gorgonio Wilderness Area and Pinnacles significant at the 0.05 level and slopes of -0.19 dv/yr and -0.21 dv/yr, respectively; Redwoods National Park and Point Reyes National Seashore are significant at the 0.1 level with slopes of -0.13 dv/yr and -0.21 dv/yr, respectively. On the Colorado Plateau Canyonlands National Park (-0.16 dv/yr) and Petrified Forest National Park (-0.15 dv/yr) are significant at the 0.1 level. Weminuche Wilderness Area in the Central Rockies has a slope of -0.13 dv/yr and is significant at the 0.05 level. Mount Rainier National Park in the Cascades and Glacier National Park in the Northern Rockies are significant at the 0.1 level and have slopes of -0.16 dv/yr and -0.09 dv/yr. These are consistent with the findings of Iyer et al.[50] who found that the proportion of high concentration days decreased for fine mass at many western sites and Acadia, which seems to have been driven by decreased proportions of high concentration days for sulfur, elemental carbon, and organic carbon.

Trends for $PM_{2.5}$ in the worst days (Table 5) essentially follow the deciview trends with few exceptions. Only four sites have slopes for $PM_{2.5}$ that are opposite in sign for deciview; one of which is Badlands, the others are Crater Lake, Shenandoah, and Washington, D.C. Three of these sites have negative slope estimates with the estimate for Washington, D.C., being positive; however, the slopes are highly insignificant. Five additional sites with positive slopes are Big Bend, Chiricahua, Great Smoky Mountains, Guadalupe Mountains, and Yosemite none of which were significant. The remaining 24 sites all have negative slopes of which seven sites had significant slopes. Acadia, Canyonlands, and Glacier are significant at the 0.1 level and of slopes of –0.18 $\mu g/m^3$, -0.12 $\mu g/m^3$, and -0.11 $\mu g/m^3$, respectively. Mount Rainier, Petrified Forest, Pinnicles, and San Gorgonio have slopes of –0.20 $\mu g/m^3$, -0.11 $\mu g/m^3$, -0.21 $\mu g/m^3$, and 0.37 $\mu g/m^3$, respectively, and are significant at the 0.05 level of probability.

The median days (Figure 8, Table 6), group 50, have negative slopes for deciview at all sites, eighteen of which are significant. In the east, Shenandoah (-0.13 dv/yr) is significant at the 0.1 level and Acadia at the 0.05 level has a slope of -0.15 dv/yr. In the west, the sites with insigificant slope are Big Bend, Guadalupe Mountains, Mesa Verde, Grand Canyon, Bridger, Yellowstone, San Gorgonio, Yosemite, Crater Lake, and Mount Rainier. Four sites are significant at the 0.1 level: Denali, Chiricahua, Tonto, and Bryce Canyon. The remaining 12 western sites have negative slopes and are significant at the 0.05 level.

As for deciview, the median days (Table 6) have all negative slope estimates for $PM_{2.5}$, of which only six sites (Crater Lake, Great Smoky Mountains, Mesa Verde, San Gorgonio,

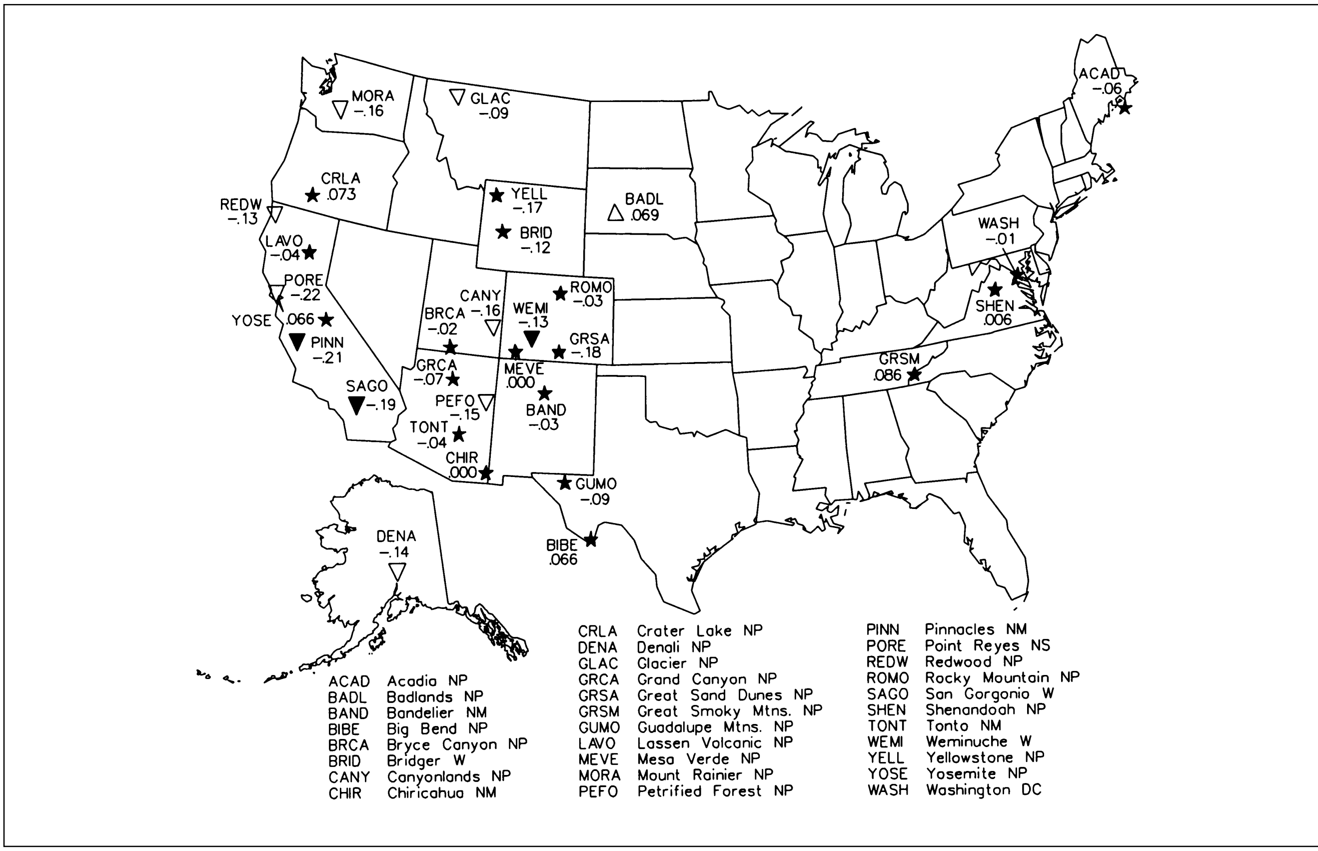

Figure 7. United States maps showing the Theil slope estimates and significance for deciview at the 30 IMPROVE sites with data beginning in March 1988 for the worst 20% days (group 90). The icons indicate site location. A star indicates non-significance, an open triangle (up or down) indicates significance at the 0.1 level of probability, the solid triangle indicates significance at the 0.05 level.

Washington DC, and Yosemite) were insignificant. Big Bend, Chiricahua, Guadalupe Mountains, and Mount Rainier with slopes of -0.05 μg/m^3, -0.05 μg/m^3, -0.06 μg/m^3 and -0.09 μg/m^3, respectively, were significant at the 0.1 level. The remaining 20 sites were significant at the 0.05 level or better and had slopes ranging from -0.05 μg/m^3 at Denali to the second most negative at Glacier, at –0.13 μg/m^3. The most negative slope occurred at San Gorgonio at –0.23 μg/m^3 but is just barely insignificant at the 0.1 level.

The cleanest days (Figure 9, Table 7), group 10, has all negative slope of which 13 are significant at the 0.05 level and four at the 0.1 level. In the east, only Acadia with a slope of -0.24 dv/yr is significant at the 0.05 level, Washington, D.C., Shenandoah, and Great Smoky Mountains are insignificant. In the intermountain west, which includes the central Rockies, the Colorado Plateau, and the Sonoran regions, all sites except Chiricahua and Weminuche have significant negative slopes. Grand Canyon, Mesa Verde, and Bandalier are significant at the 0.1 level and have a slope of -0.1 dv/yr, -0.15 dv/yr, and -0.17 dv/yr, respectively. The remaining intermountain sites are significant at the 0.05 level and have slopes ranging from -0.33 dv/yr at Yellowstone to -0.13 dv/yr at Bryce Canyon. Pinnacles (-0.21 dv/yr), Redwoods (-0.19 dv/yr), Crater Lake (-0.17 dv/yr), and Lassen Volcanoes (-0.17 dv/yr) are significant at the 0.05 level.

All slope estimates for $PM_{2.5}$ are negative for the cleanest days (Table 7). In the east, Acadia and Great Smoky Mountains have significant slopes of –0.10 μg/m^3 and –0.08 μg/m^3, respectively, while the slopes at Shenandoah at –0.04 μg/m^3 and Washington DC at –0.1 μg/m^3 are insignificant. In the west, all sites except Big Bend, Chiricahua, and San Gorgonio have significant slopes. For this group it is notable that 15 sites are significant at the 0.01 level and include Acadia, Badlands, Bryce Canyon, Canyonlands, Crater Lake, Denali, Glacier, Grand Canyon, Great Sand Dunes, Lassen Volcanoes, Petrified Forest, Redwoods, Rocky Mountain, Weminuche, and Yellowstone with slopes ranging from –0.04 μg/m^3 at Weminuche to –0.11 μg/m^3 at Yellowstone. Three sites (Great Smoky Mountains, Point Reyes, and Yosemite) are significant at the 0.1 level, and the remaining sites are significant at the 0.05 level. These finding are consistant with Iyer et al.,[50] who found that the proportion of low concentration days increased for organics, and fine mass at most sites: however, sulfur and elemental carbon were mixed with sites showing increased proportions juxtaposed with sites showing decreased proportions.

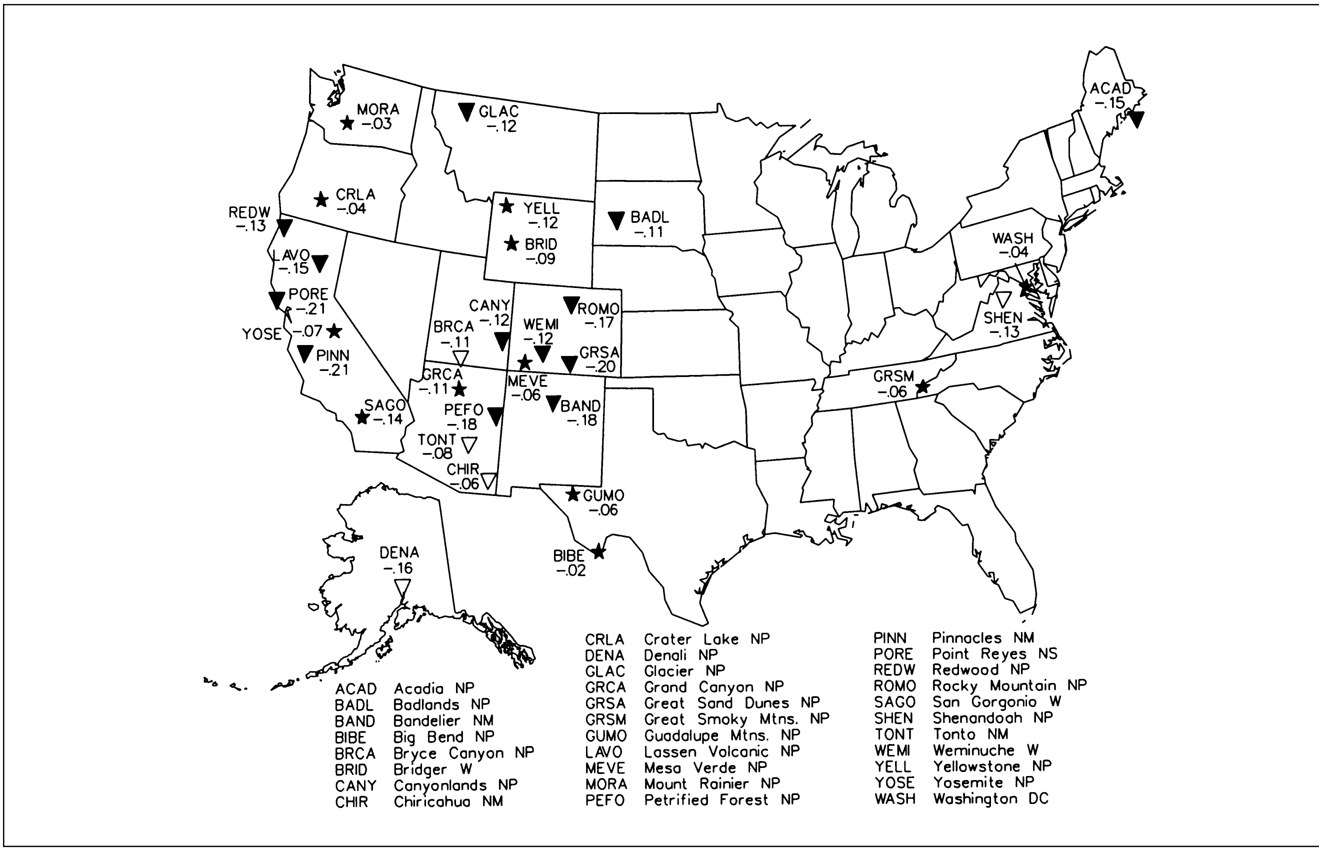

Figure 8. U.S. maps showing the Theil slope estimates and significance for deciview at the 30 IMPROVE sites with data beginning in March 1988 for the median 20% days (group 50). The icons indicate site location. A star indicates non-significance, an open triangle (up or down) indicates significance at the 0.1 level of probability, and the solid triangle indicates significance at the 0.05 level.

CONCLUSIONS

The east-west dichotomy in aerosol conditions is quite evident as shown by the pie charts (Figures 3a and 3b). There is a factor of three difference in east-west loadings of $PM_{2.5}$ with eastern aerosols being dominated by sulfate. While in the west, carbonaceous species (organic mass and elemental carbon) frequently exceed sulfate as the chief contributor to $PM_{2.5}$. Due to RH affects the east-west differences are exacerbated with a factor of 5 difference between the east and west with sulfate extinction dominating aerosol extinction. In the west, due to RH, sulfate extinction is generally on a par with extinction due to carbonaceous species. Southern California is unique in that nitrate dominates $PM_{2.5}$ and nitrate extinction dominates aerosol extinction.

A principal conclusion from this visibility trends analysis, which assesses 29 rural and one urban site with continuous data for the period 1988–1996, is that there is no significant deterioration of air quality and visibility at most monitoring locations. Annual median (group 50) and best (group 10) visibility conditions are gradually improving at about 70% of the sites based on the significance of the slope estimate for deciview. $PM_{2.5}$ is slightly better for groups 10 and 50 with about 80% of the sites showing significant improvement. The worst visibility days are showing significant improvement at about 30% of the sites for both $PM_{2.5}$ and deciview.

There may be cause for some concern, however, about the number of sites not showing steady improvements, particularly for the worst 20% days (group 90). As shown by Table 5, the ten sites that show degrading conditions (positive slopes) for either deciview or $PM_{2.5}$ are Badlands, Big Bend, Chiricahua, Crater Lake, Great Smoky Mountains, Guadalupe Mountains, Mesa Verde, Shenandoah, Yosemite, and Washington, D.C. Of these ten sites, only Big Bend, Great Smoky Mountains and, Yosemite had positive slopes for both deciview and $PM_{2.5}$. Two of the sites, Chiricahua and Mesa Verde, had slopes of zero for deciview with the slopes for $PM_{2.5}$ positive at Chiricahua and negative at Mesa Verde. For the remaining five sites all slopes were negative. However, for all of these ten sites, only the positive slope estimate for deciview at Badlands was significant.

The 20 remaining sites from the worst visibility days show improvement in deciview and $PM_{2.5}$ (negative slopes) two groups emerge; those sites showing significant improvement and those sites with no significant improvement. Eight of the sites that have significant slope

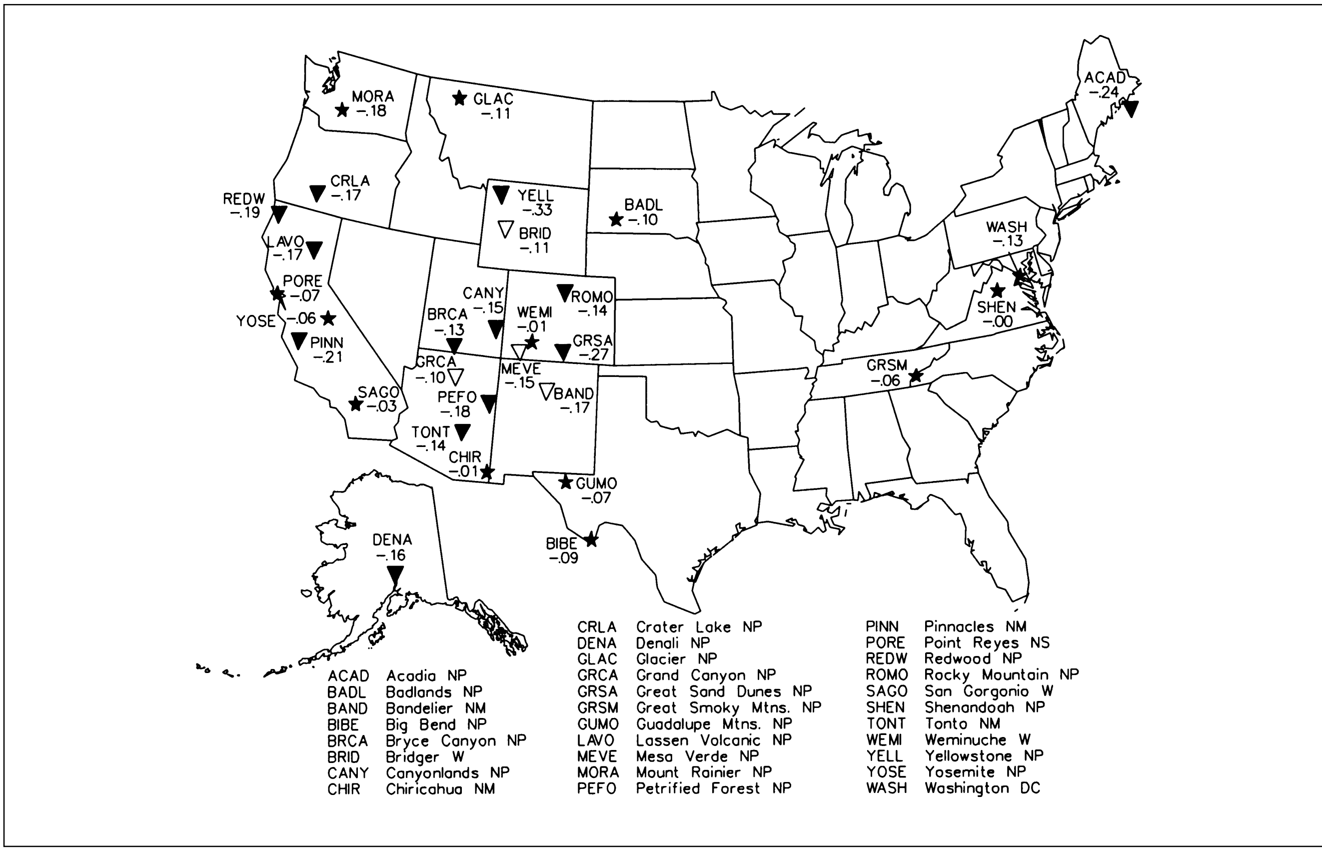

Figure 9. United States maps showing the Theil slope estimates and significance for deciview at the 30 IMPROVE sites with data beginning in March 1988 for the best 20% days (group 10). The icons indicate site location. A star indicates non-significance, an open triangle (up or down) indicates significance at the 0.1 level of probability, the solid triangle indicates significance at the 0.05 level.

estimates for both deciview or $PM_{2.5}$ are Canyonlands, Glacier, Mount Rainier, Petrified Forest, Pinnacles, Point Reyes, Redwood, and San Gorgonio. Three additional sites significant only for either deciview or $PM_{2.5}$ are Denali and Weminuche for deciview and Acadia for $PM_{2.5}$. The nine sites showing insignificant improvement for the worst visibility days are Bandelier, Bryce Canyon, Bridger, Grand Canyon, Great Sand Dunes, Lassen Volcanoes, Rocky Mountain, Tonto, and Yellowstone. While annual data show that conditions are not improving in the three rural eastern sites, it is important to note that they are expected to have fairly substantial improvements in visibility (2-4 deciviews) by the year 2010 due to sulfate reductions required under the acid rain program.[46]

In regard to the median values (group 50) as shown in Table 6, it appears visibility conditions are gradually improving with about 80% of the sites showing significant slopes either deciview or $PM_{2.5}$; moreover, all slope estimates are negative regardless of significance. The six sites with insignificant slopes for deciview and $PM_{2.5}$ are Crater Lake, Great Smoky Mountains, Mesa Verde, San Gorgonio, Washington, DC, and Yosemite. An additional six sites, Big Bend, Bridger, Grand Canyon, Guadalupe Mountains, Mount Rainier, and Yellowstone had significant slopes for only $PM_{2.5}$. The remaining 18 sites had significant slopes for both deciview and $PM_{2.5}$.

Trends for group 10 (Table 7), or best visibility days, appear to show that, with the exception of thirteen sites in the case of deciview and six sites in the case of $PM_{2.5}$, most sites are showing significant improvements in visibility conditions. All sites had negative slopes for both deciview and $PM_{2.5}$. In other words, the clean days are getting cleaner.

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DISCLAIMER

The assumptions, findings, conclusions, judgments, and views presented herein are those of the authors and should not be interpreted as necessarily representing official National Park Service policies.

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