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Influence of particulate size on statistical studies of visibility at California regions

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RESUMEN

Se presenta un análisis de regresión lineal múltiple de partículas, gases y datos meteorológicos para diferentes sitios en California. Los resultados del análisis de regresión proporcionan algunas ideas sobre los procesos que deterioran la visibilidad. Un resultado notable es la poca o ninguna relación entre la visibilidad y el abundante componente de azufre con diámetro menor que 0.65 μm. Se concluye que la visibilidad pobre se debe al crecimiento de las partículas arriba de 0.65 μm; crecimiento que es favorecido por las humedades altas pero que también puede reflejar las fuentes elementales del azufre. Este estudio ha demostrado el papel importante del tamaño de las partículas en la extinción atmosférica. Aquellas condiciones que favorecen la formación o crecimiento de partículas en su parte óptica efectiva en forma de acumulación, están conectadas con la reducción de la visibilidad. Estos resultados están en buena concordancia con la teoría de Mie, y permiten mejor acuerdo entre la teoría y los experimentos que los obtenidos sumando toda la masa abajo de 2.5 μm o incluso componentes químicos claves en el régimen de menos de 2.5 μm.

ABSTRACT

A multiple linear regression analysis of particulate, gas, and meteorological data is presented for different sites in California. The results of the regression analysis provide some insight into visibility-impairing processes. One remarkable factor is little or no association between visibility and the abundant sulfur component with diameter less than 0.65 μm. One must conclude that it was the growth of particles above 0.65 μm that caused the poor visibility; growth that was favored by the higher humidities but that may also reflect the ultimate sources of the sulfur. This study has shown the key role played by particulate size in atmospheric extinction. Those conditions that encourage the formation or growth of particles in the optically effective part of the accumulation mode are linked to reduced visibility. These results are in good agreement with Mie theory, and allow a much better match between theory and experiments than those gained by summing all mass below 2.5 μm or even key chemical constituents in the less than 2.5 μm regime.

Introduction

The reduction of visibility due to the turbidity of polluted air continues to be one of the most evident indicators of atmospheric degradation, and thus a constant source of societal concern. In order to design a strategy for the protection of visibility, one would like to know the relative contributions to light extinction by the major aerosol constituents. In the absence of the detailed aerosol data necessary to estimate these contributions from optical theory, it is common to infer them from the empirical relationship of extinction to aerosol composition. While the absorption

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of light by gaseous nitrogen dioxide ($N0_2$) can cause visibility degradation, more important is the scattering of light by particles. In particular, particles between 0.3 and 1 μm in diameter have both long residence times and high light scattering per unit mass (according to Mie theory), since they are about the same size as the wavelength of visible light. As a consequence of the fairly long residence time that small particles enjoy in the atmosphere, the duration of poor visibility is usually determined by meteorological conditions and ventilation.

A number of studies have employed multivariate techniques to examine relationships between atmospheric extinction and/or scattering and various atmospheric pollutants. Henry and Hidy (1979), and Pitchford *et al.* (1981) used factor analysis and principal component analysis to develop insight into visibility-particulate relationships. Others, such as Trijonis and Yuan (1978), White and Roberts (1977), Barone *et al.* (1978), Cobourn and Husar (1982), and White (1986) have examined relationships between atmospheric extinction (or scattering) and atmospheric particulate matter through the use of multiple regression analysis. Barone *et al.* (1978) used a multiplicative regression model to interpret the relationship between visibility reduction, pollutants, and meteorological conditions at four California sites in summer 1973. It appears that there were some problems with early data. Thus, this study is based on data from the summer 1975 through the summer 1977 at six California sites, and we used a multiple linear regression model rather than a multiplicative model.

Mie scattering theory predicts that knowledge of both particle size and complex index of refraction are necessary to calculate optical extinction. However, few studies of visibility include simultaneous measurements of particle size and chemical composition. The latter factor is presumably important in the index of refraction. The situation is inherently complex because of the large number of parameters that may be implicated in visibility reduction; thus, one is required to make either numerous detailed measurements by size and composition or to accept potentially erroneous limitations in the data set that may foreordain the conclusion. One data set that at least partially surmounts these limitations was collected in California in the period from 1973 through 1977; it included three size fractions and elemental analysis. When these 24-hour values were combined with weather parameters and gaseous pollutants on a daily basis for each 90-day season, one had a data set consisting of over 5,000 measurements from which one could extract associations with visibility.

Another comprehensive study (Trijonis, 1982) was conducted of visibility in California using prevailing visibility measurements at 67 weather stations in conjunction with data on particulate matter concentrations and meteorology. As part of that study, visibility/aerosol relationships were analyzed using 3 to 8 years of data at 12 California locations having nearby airport data and Hi-Vol data. The analysis was based on statistical regression equations that relate daytime average extinction to daily averages of sulfates, nitrates, the remainder of total suspended particulate (TSP), and relative humidity.

The purpose of this paper is to attempt to combine these two California studies in such a way as to isolate the role of particle size and chemical composition in statistical studies of California visibility. One key question is to what extent size information is needed to clarify and extend statistical studies of visibility. In this regard, California also has an advantage of rather radically different meteorological condition even in the same season, due to the effect of topography on California weather.

Data description

The particulate data used in this study were available through a program operated by the University of California, Davis, in conjunction with the California Air Resources Board. Two-stage Lundgren-type rotary drum impactors with after-filters were used to collect twenty-four hour aerosol samples in three size ranges: 0.1-0.65 μm (stage 3), 0.65-3.6μm (stage 2), and 3.6-20 μm (stage 1) (effective aerodynamic diameters at unit density). The rotary drum stages were coated to minimize bounce-off problems (Wesolowski *et al.*, 1973). The final stage of the sampling device utilizes a 0.4 μm Nuclepore filter yielding a 95% collection efficiency in 0.1-0.65 μm size range. A full description of the collection efficiency, calibration, and analysis from Lundgren-type impactors used in this study is included in Flocchini *et al.* (1976) and Cahill *et al.* (1976). All samples were analyzed for elemental content by Particle Induced X-ray Emission (PIXE) (Cahill *et al.,* 1976), using the UC Davis 76" isochronous cyclotron. All elements heavier than sodium can be detected with this method at sensitivities typically of a few nanograms per cubic meter when interferences are not present (Flocchini *et al.,* 1972).

The visibility data used in this study consisted of routine prevailing visibility measurements made at weather stations (usually airports) . Because daytime and nighttime visibility measurements are often incompatible, and because the daytime data are usually of higher quality (Trijonis and Yuan, 1978; Trijonis, 1979), only daytime observations were employed in this study. The relative humidity data are also obtained from the same airport as the visibility observations. The visibility and relative humidity data represent averages of four daytime recordings, while the particulate data represent complete 24-hour averages.

Data for gaseous pollutants such as nitrogen dioxide and sulfur dioxide were obtained from the California Air Resources Board (ARB) and local air pollution control district gas monitoring programs. These data were collected at the same stations as were particulate data.

Site descriptions

The sites were selected in such a way as to represent aerosol typical of larger areas throughout California. From north to south they are: Sacramento, Oakland, San Jose, Visalia, Bakersfield, Los Angeles, Riverside, Lennox, La Habra, Los Alamitos, Anaheim, and San Diego (Figure 1). The locations cluster in the populated areas of the state. Although data availability was an important consideration, these site were selected because they contain a variety of topographic and meteorological conditions. The major weather types include the cool, humid coastal sites such as the San Francisco Bay area (Oakland, San Jose); the warmer coastal basins, exemplified by the Los Angeles basin (Los Angeles, Los Alamitos, Riverside); and the interior valleys with hot, dry summers (Sacramento, Bakersfield, Visalia). Moreover, each site experiences periods of reduced visibility and all sites are affected by a variety of pollutant sources. Bakersfield and Sacramento are inland sites whose primary sources are non-industrial. Automotive and soil derived particles provide a major fraction of the aerosol. Los Angeles and Los Alamitos are the coastal cities in Southern California surrounded by many municipal and industrial pollution sources that add to the automotive and soil components to create a more complex urban atmosphere.

This study is based on data from the summer (July through September) 1975 through the summer 1977, though not continuously at all sites. It is necessary to note that these data cover a variety of topographic and meteorological conditions throughout diverse regions in California.

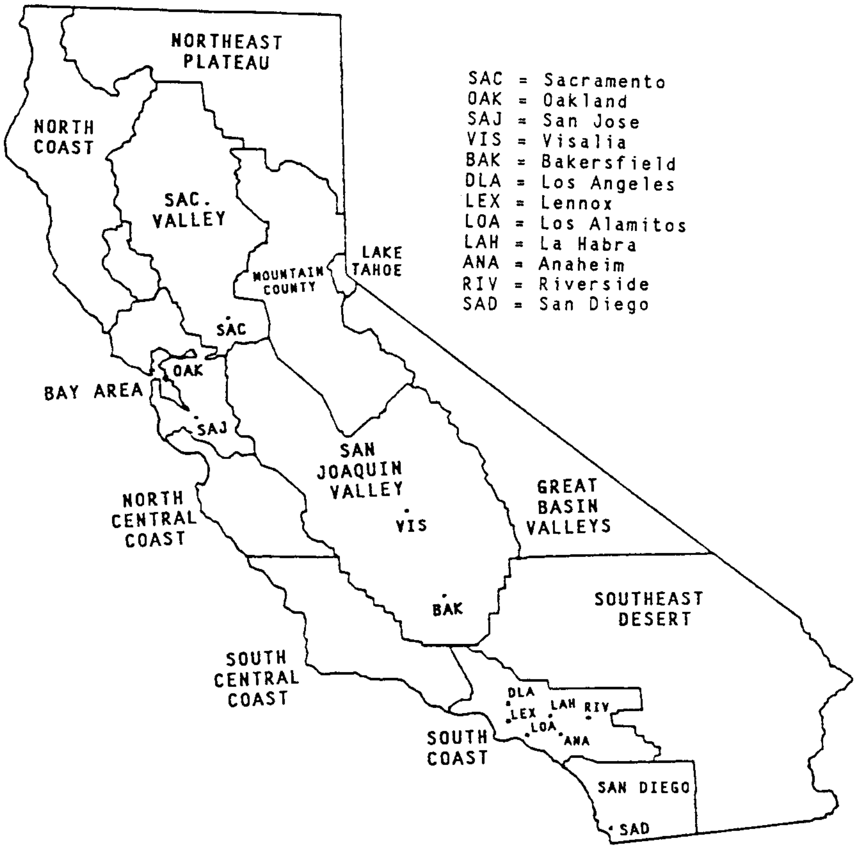

Fig. 1. Map of the California indicating the sampling sites for particulate and gaseous pollutants.

Statistical procedures

The statistical technique that allows for determining the light scattering efficiency of each component of the particulate concentration is multiple linear regression. Regression analysis yields a quantitative relationship between a dependent variable such as light scattering coefficient and independent variables such as elemental composition.

In general, multiple linear regression of a dependent variable y on independent variables $x_1,\dots,x_n$ determines coefficients $a_1,\dots,a_n$ which minimize the mean square error. This approach does not yield information on the relationship between independent variables, but only defines the relationship between the dependent variable and each independent variable. Although, in general, there are no restrictions on what species can be used in a multiple regression analysis, a highly directed approach is usually taken where specific features are selected to represent specific sources, for example Pb for automotive exhaust. This is usually the case because of the large number of combinations and permutations that could otherwise be involved. Whatever species are used, it is essential that they are independent. The linear model considered here is

$$Y = a_o + \sum_{i=1}^{n} a_i x_i + E \tag{1}$$

where the E denotes the error, and a_o is the intercept. If the linear model is physically appropriate, then the coefficients a_i can be determined with arbitrary accuracy by taking enough samples, even though uncertainty in the relationship may exist for individual samples. The variance σ and the errors corresponding to the various $Y's$ are assumed to be independent. The level of significance of the coefficients can be determined by using a "t" test. The proportion of total variation in Y accounted for by the regression model is given by the coefficient of determination, R^2.

$$R^2 = \frac{\text{Sum of Squares Regression}}{\text{Sum of Squares Total}}$$

In this regression analysis, the dependent variable would be the light scattering coefficient (b_{scat}) which is calculated from the Koschmieder formula, b_{scat}= K/ visual range. The constant is usually chosen to be K = 3.9 or K = 2.9, depending on whether one assumes a 2% or 5% contrast detection threshold for the observer. In the derivation of the Koschmieder formula certain assumptions were made: the extinction coefficient is constant along the path of sight (this also means a horizontal path of sight and neglecting the curvature of the Earth); the amount of the light scattered by a volume element is proportional to its volume and extinction coefficient, and also is constant along the path of sight; the target is absolutely black, with horizon as background; sky is cloudless. Despite these restrictions the formula has been widely used for various purposes such as meteorological records, airport visibility, or environmental studies, and it has been applied to calculations of visibilities from measured extinction or scattering coefficients or vice versa. The variables assumed to be independent in regression analysis were the concentration of atmospheric aerosols (by size and elemental composition), the relative humidity, and the concentration of gaseous pollutants (NO_2, SO_2). The regression coefficient corresponding to each independent variable can be interpreted as the extinction efficiency of that variable. A more complete discussion of multiple linear regression analysis is available elsewhere (Draper and Smith, 1966).

Data analysis and interpretation

The seasonal variations of sulfur concentration and light scattering coefficient for selected sites are shown in Figure 3. Different sources of sulfur have been found to have different size distributions. For example, Ouimette (1980) and Friedlander (1980) have observed two distinctly different sulfur size distributions in California, one in coastal regions with an aerodynamic mass median diameter (MMD) >0.5 μm thought to be the result of SO_2 conversion in fogs in the droplet phase and the other in the desert area with MMD ~ 0.2 μm ascribed to homogeneous gas-phase SO_2 conversion reactions. The role of particle size should be mentioned at this time. Sulfur shows a considerable seasonal variation. The winter sulfur size appears to be in the accumulation mode and quite fine. In summer, however, sulfur particles grow in size up to the very upper edge of the accumulation mode. During this period, the amount of sulfate more than doubles while the trace elements associated with oil combustion, vanadium and nickel, remain at their winter size profile. Thus, the primary combustion sulfates have a constant accumulation mode size profile, and the new summer sulfate mode is a secondary aerosol conversion. The summer sulfate aerosol appears hygroscopic, growing in size and accelerating gas to particle conversion via the aqueous state. Such a model would also predict an important role for relative humidity when gas-to-particle conversion is occurring in the higher oxidant, higher temperature summer periods (Trijonis, 1982).

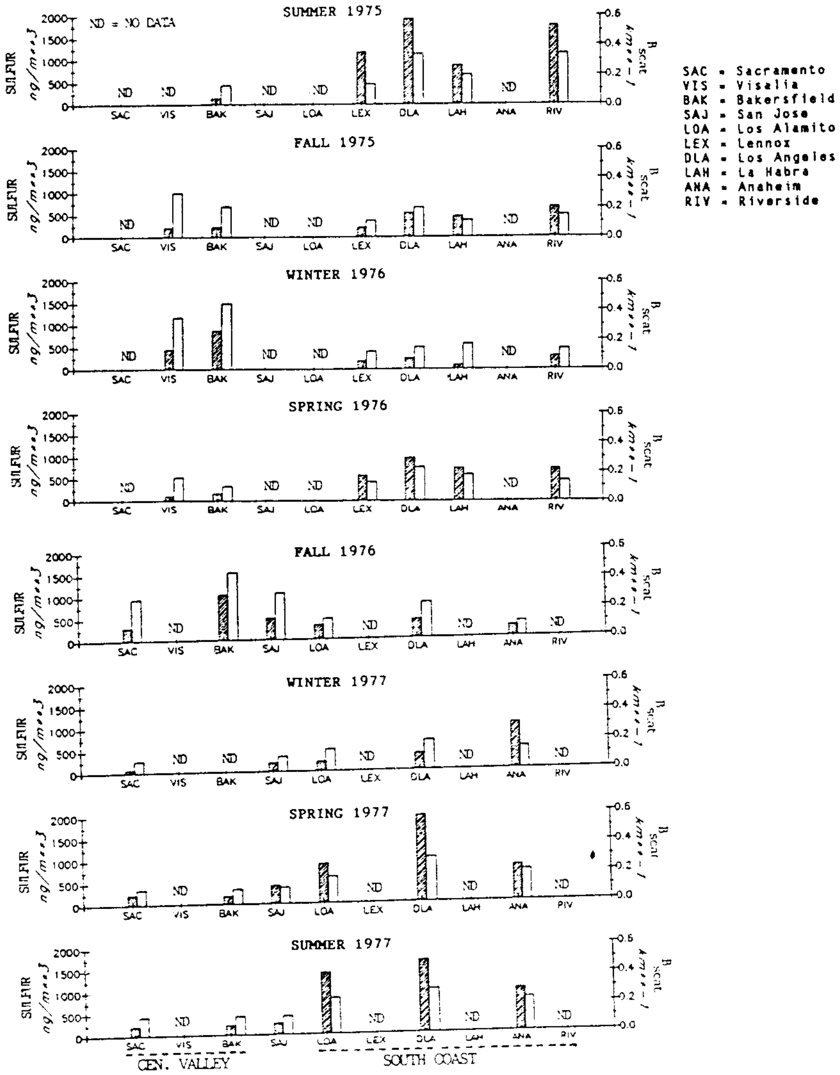

Fig. 2. Seasonal variations of sulfur concentrations, and scattering coefficients by sites, Summer (July, Aug., Sept.), Fall (Oct., Nov., Dec.), Winter (Jan., Feb., Mar.), Spring (Apr., May, June). The sulfur concentration is cross hatched.

Generally, Figure 2 depicts a high degree of association between the sulfur concentration and the light scattering coefficient, calculated from the Koschmieder formula, for all of the sites (there was no measurement of scattering coefficient with a nephelometer). The experimental points yield an average bivariate correlation coefficient of 0.76 for the South Coast Air Basin, and 0.62 for the Central Valley. Figure 2 also shows high summer-time sulfur concentration within the South Coast Air Basin, especially in Los Angeles. More intense sunshine, stronger inversion ceilings, and greater inland penetration of moist air all serve to increase sulfur concentration and other photochemical aerosols during summer. Los Angeles area also tends to have relatively high values in the spring. The relatively higher values in the summer and spring might be related to the higher transformation rate during these seasons. The relatively higher values in the summer might also be related to the lower magnitudes of the average directional transport wind components during this season. The airflow patterns in the South Coast Air Basin show that the westerly flow is the most frequent in the periods April to June and July to September. This type of airflow allows pollutants to build up in the industrial and business areas and then move inland with the sea breeze in the afternoon. Central Valley has relatively higher values in the winter, and lower values in the summer. Concerning the different trend for sulfur concentration in the Southern California region and the Central Valley, Flocchini *et al.* (1978) mentioned that the mechanism for the increased proportion of stage 2 sulfur in Bakersfield appears to be different from that in Los Angeles. The oxidizing atmosphere of Los Angeles may result in the early summer peak in the intermediate range whereas the increase in relative humidity and lower mixing depth at Bakersfield during the winter may account for the peak in the intermediate range.

The results of the multiple regression analysis

In this study the relationship between visibility reduction, atmospheric pollutants and relative humidity is examined by using multiple linear regression analysis. Meanwhile, in order to find the relationship between aerosols and visibility, both the size distribution and the elemental composition of particulate aerosols are considered. It is necessary to note that there are several limitations to the use of regression models for quantifying visibility/aerosol relationships. A detailed discussion of how the limitations may have affected the results of a regression model was presented by Trijonis (1982). Although the regression models are subject to several limitations, the conclusions resulting from these models have proven to be very reasonable.

Tabla 1. Significant beta coefficients for selected sites (July-September).

Variable	Sacramento (1977)	Bakersfield (1977)	San Jose (1977)	Los Angeles (1977)	Los Alamitos (1977)	Riverside (1975)
Na-2	-.37	-.04	-.13	-.22	-.23	-.18
Si-2	-.39	-.33	****	****	.09	****
S-2	.41	.55	.53	.82	.54	.33
K-2	.43	.31	-.66	****	-.31	****
Pb-2	****	****	-.36	****	****	****
Na-3	****	****	****	****	****	****
S-3	.35	.26	.29	****	.20	.21
K-3	****	-.23	.58	-.12	****	****
Pb-3	****	****	****	.32	****	-.34
NO_2	****	.54	.59	.24	——	****
SO_2	****	****	——	.52	****	——
RH	.23	.19	.53	.45	.37	.46
Multiple R^2	.77	.68	.81	.75	.70	.71

*** * * indicates the beta coefficient is not significant at the 0.1 confidence level for the t-test**
2 indicates intermediate size particles 0.65-3.6 μm
3 indicates fine particles 0.1-0.65 μm
- indicates no data

The beta coefficients for the selected California sites are reported in Table 1. The beta coefficient is a standardized measure (i.e. dimensionless) of the effect that an independent variable (e.g. sulfur) has on the dependent variable (b_{scat}) while holding constant all other independent variables in the regression equation. The beta coefficient relys on the amount of variance in the variables, but not their magnitude. The beta coefficient computed for each variable at each site is a measure of the relative importance of that variable in determining b_{scat} (i. e. the larger the absolute value of the beta coefficient the greater the effect of the independent variable on the dependent variable).

In order to determine which elemental aerosols were associated with specific sources, bivariate correlations and element-to-element ratios were computed for all sites within each size range. These data were used to determine which elements occurred on similar particles or had similar origins. Since many variables appear to originate from the same source, the dimensionality of the regression equation can be greatly reduced by choosing a single representative variable from each source and using it in the regression equation. In this way, the number of highly correlated variables from the regression equation is reduced which in term reduces problems of multicolinearity. It should be mentioned that aerosol particles greater than 3.6 μm were excluded from the analysis since the residence time for these particles in the atmosphere is short and previous studies indicate that these large particles play no important role in visibility reduction.

Table 1 shows the significant beta coefficients for selected sites. The Sacramento area exhibits a relatively strong dependence of b_{scat} on two elements (sulfur and potassium) in the size region from 0.65 to 3.6 μm. These elements are bivariably unrelated to one another, which means they would not occur predictably on the same particle and therefore probably originate from two separate and unique sources. Potassium also did not exhibit very high bivariate correlations with those aerosol components generally labeled soil derived, i. e. Si, Al, and Fe, thus, it might have originated mostly from agricultural burning. The effect of sulfur in the 0.65–3.6 μm size range on visibility reduction at Bakersfield (summer 1977) is stronger than that for Sacramento, but fine potassium aerosols of soil derivation or agricultural burning were relatively less important. The positive beta coefficient (0.54) associated with nitrogen dioxide at this site may be acting as a surrogate for vehicular activity (particles) and that Bakersfield is dominated by two sources, crude oil production and vehicles. At San Jose, visibility is related to the relative humidity. This result is expected since the role of water vapor in visibility reduction is well known. A relatively strong positive beta coefficient associated with sulfur at this site suggests again that this is the significant visibility limiting aerosol. The positive beta coefficient associated with nitrogen dioxide is relatively similar in magnitude to the beta for this variable at Bakersfield (summer 1977). The negative beta coefficient for lead is not very clear, but may be related to wind direction.

At the Los Angeles site, sulfur aerosols in the 0.65- 3.6 μm size region, with a beta coefficient of 0.82, was the most important variable related to visibility reduction. Humidity, with a beta coefficient of 0.45, was also a significant factor. The sulfur aerosols may be due to conversion of SO_2 to sulfate, as indicated by significant beta coefficient associated with SO_2 at this site. The positive beta coefficient associated with nitrogen dioxide at this site indicates that NO_2 is responsible for light absorption in the visible region of spectrum. As proposed by Trijonis (1982), the nitrate contributions to the light extinction are larger in the Central Valley than in the Southern California. It should be mentioned that the visibility/nitrate relationships are very uncertain because of statistical colinearity problems and because of measurement difficulties for nitrate. The positive beta coefficient for fine lead can be related to automotive exhaust in the Los Angeles area. The results of the regression models for Los Alamitos and Riverside also indicated the dominant role of sulfur aerosols, primarily in the 0.65-3.6 μm size range, in visibility impairment. At the Riverside site, visibility reduction was most strongly related to relative humidity, as indicated by the strong positive beta coefficient associated with this variable. The effect of humidity on visibility reduction was also significant at Los Alamitos. The negative beta coefficient for sodium in the 0.65-3.6 μm size region may be related to cooling sea breezes which provide significant ventilation and improved visibility at most of the southern California sites. Figures 3 through 5 show the observed and estimated light scattering coefficients for the selected sites. The scatter plots of light scattering coefficient are shown in Figures 6 and 7 .

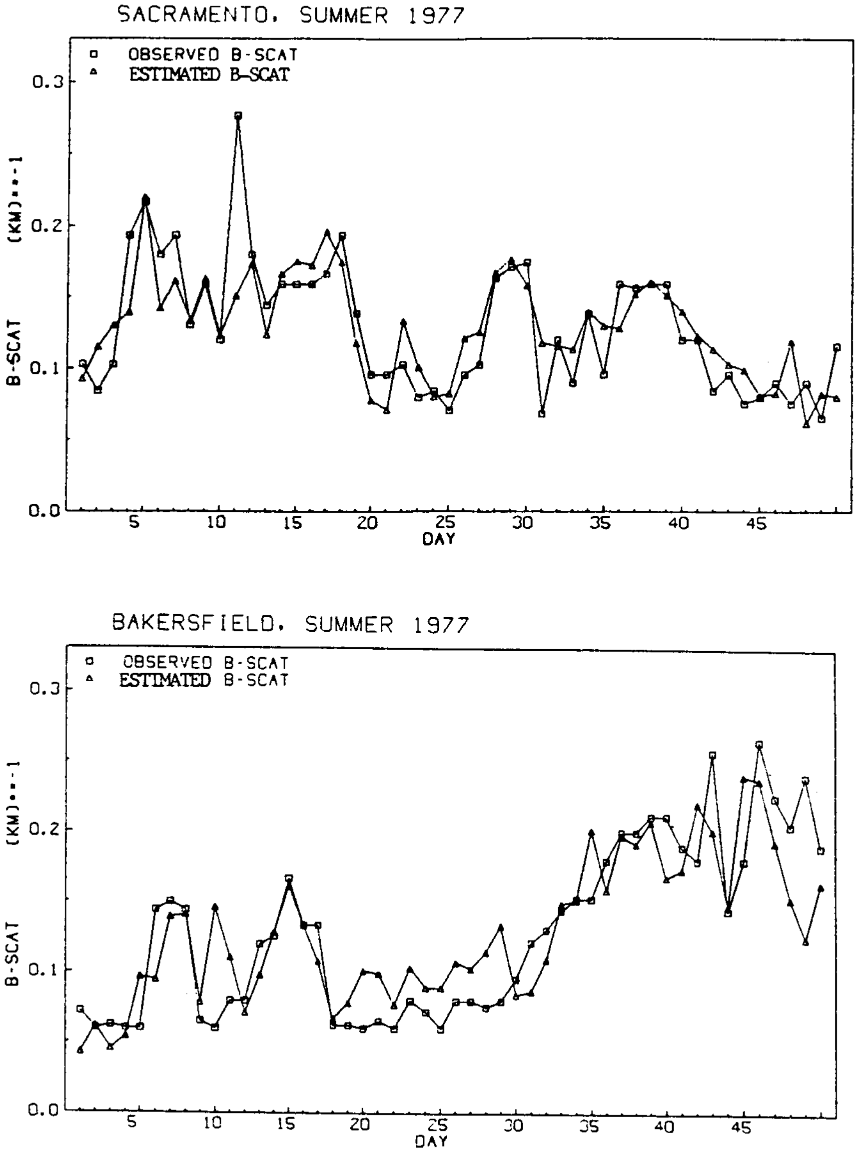

Fig. 3. Observed and estimated values for scattering coefficients (km^{-1}) at Sacramento and Bakerfield.

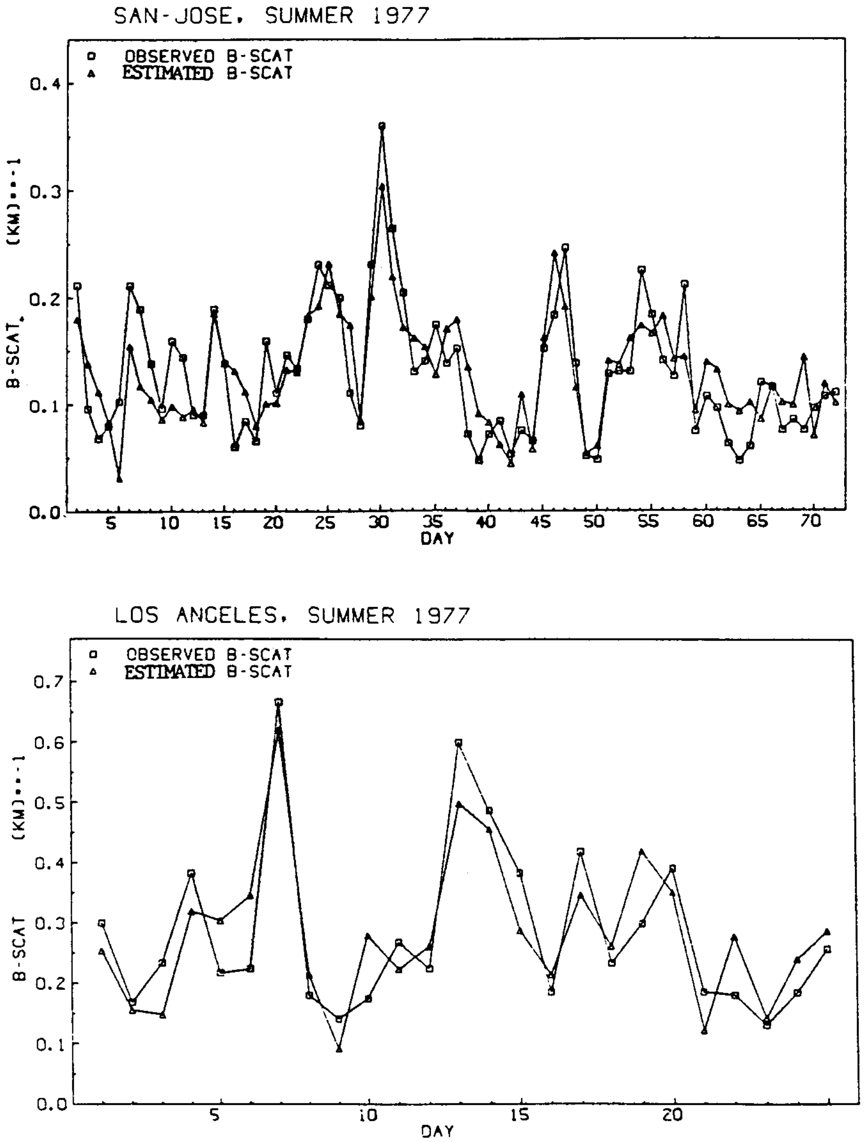

Fig. 4. Observed and estimated values for scattering coefficients (km^{-1}) at San Jose and Los Angeles.

Although the data presented here are not definitive, they provide some insight into visibility-impairing processes. One remarkable factor is little or no association between visibility and the abundant sulfur component with diameter less than 0.65 μm. One must conclude that it was the growth of particles above 0.65 μm that caused the poor visibility, growth that was favored by the higher humidities but that may also reflect the ultimate sources of the sulfur. Correlation coefficients between sulfur and the metals vanadium and nickel are very strong in the fine fraction, and the ratios are close to those in fuel oil. The correlations are weak and the ratios are sharply

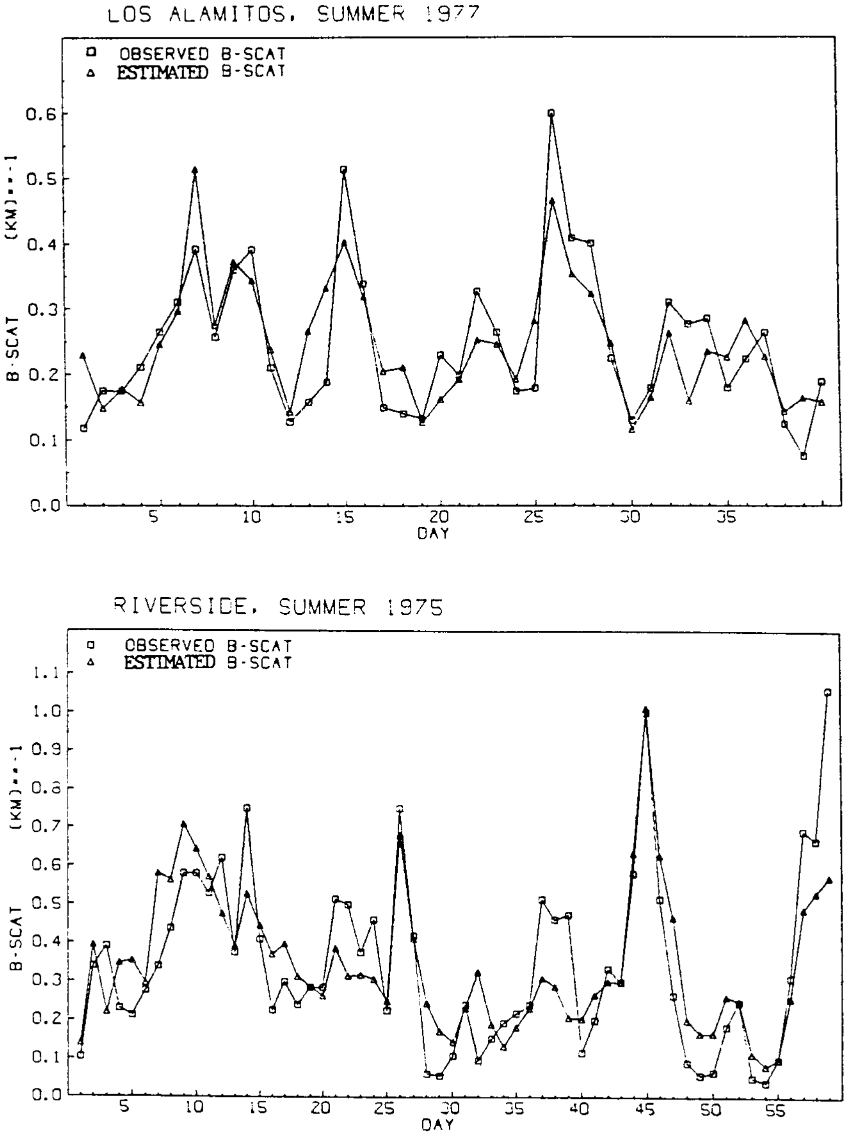

Fig. 5. Observed and estimated values for scattering coefficients (km^{-1}) at Los Alamitos and Riverside.

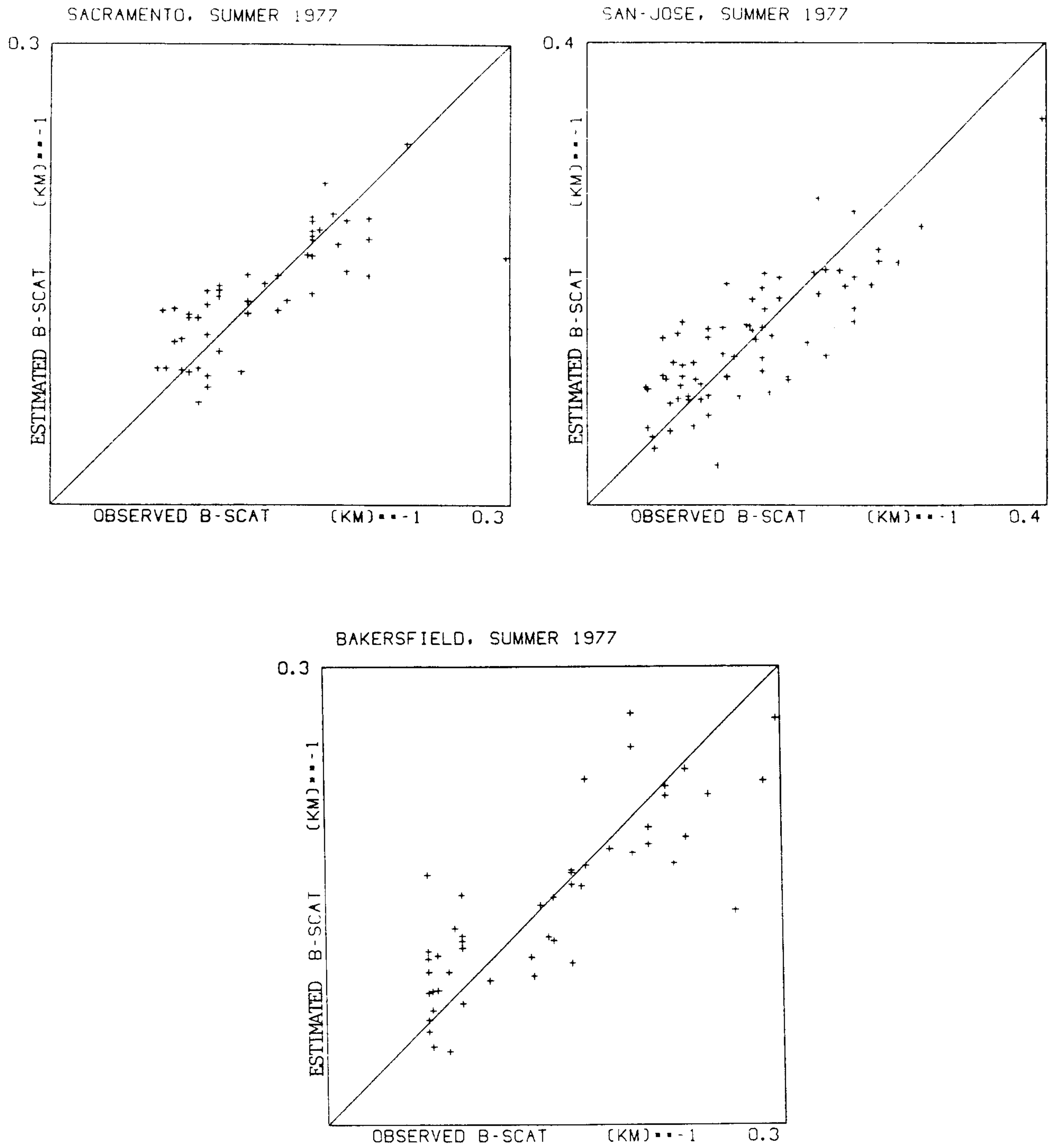

Fig. 6. Scatter plots of B_{scat} (km^{-1}) for three sites.

depressed for the metals in the intermediate size particles. Table 1 shows that the larger diameter sulfur particles are mainly present in the Los Angeles area in summer. Thus, one can associate the coarser particles with photochemical conversion of SO_2 to sulfate, whereas the finer particles appear to be direct, primary emission of particles from oil combustion. The fine particles may, in fact, be much finer than 0.65 μm; thus, their ability to scatter light is greatly reduced. The results of this study can explain the observed geographical patterns in sulfate extinction efficiency.

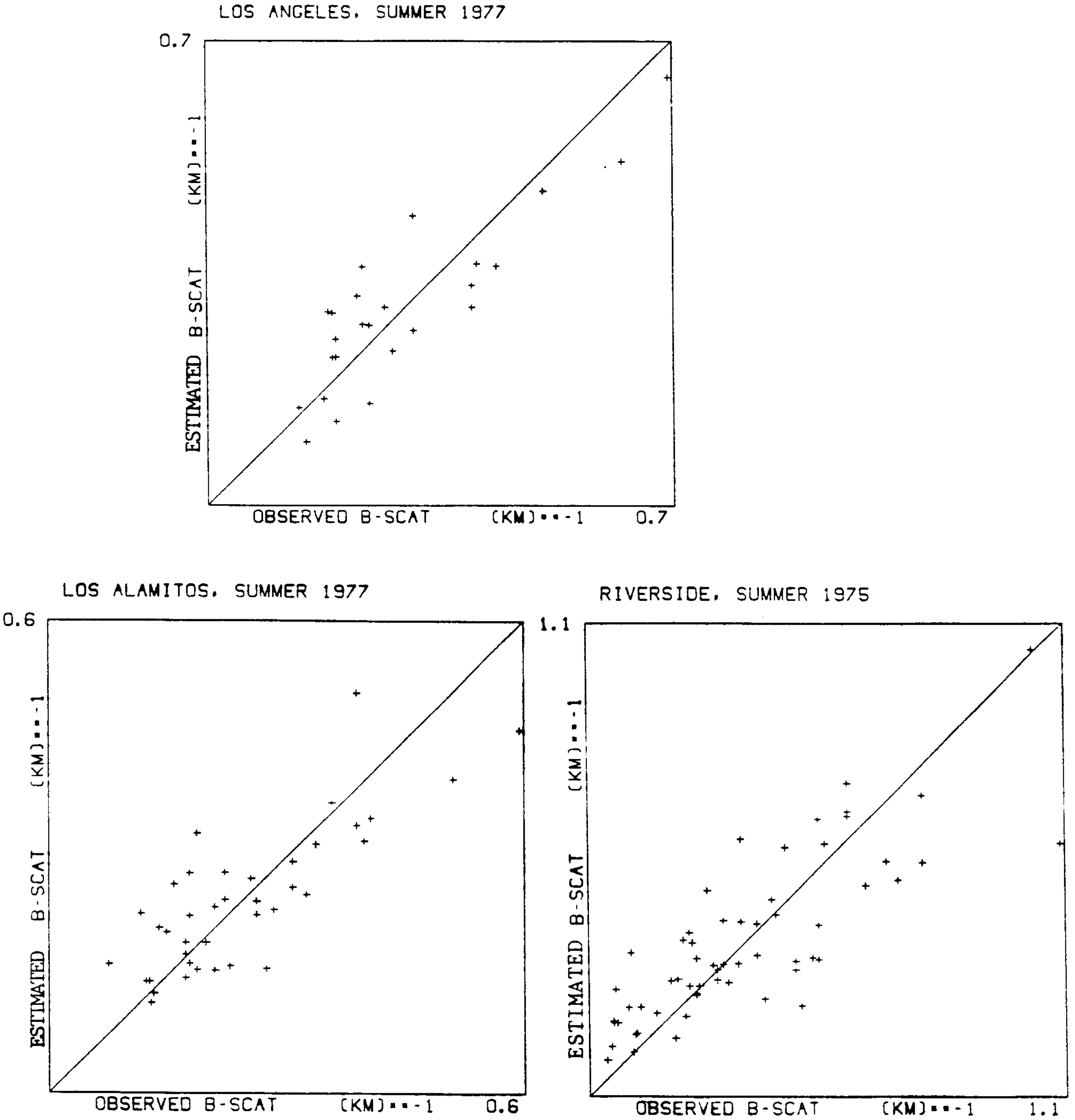

Fig. 7. Scatter plots of B_{scat} (km^{-1}) for three sites.

Trijonis *et al.* (1982) plotted regional sulfate coefficients against regional sulfate size distribution, Figure 8. This graph shows that regional sulfate extinction efficiencies do correlate in the expected manner with sulfate size distribution. Thus, one may conclude that at least some of the geographical variation in sulfate extinction efficiency is related to the phenomenon of sulfate size distribution.

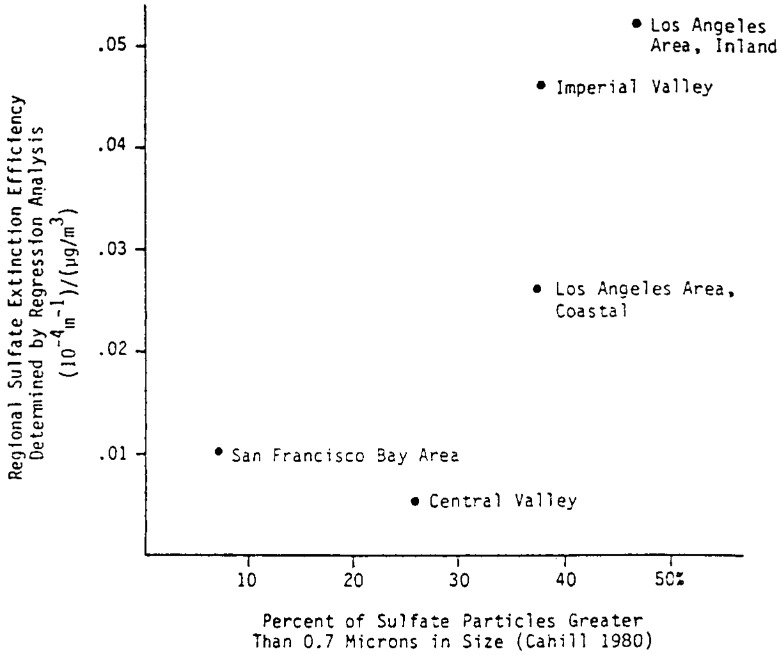

Fig. 8. Relationship between regional extinction efficiency for sulfates and sulfate size distribution. (Trijonis *et al.*, 1982).

Conclusions

A comprehensive study was conducted of visibility in California using prevailing visibility measurements made at weather stations (airports) in conjunction with data on particulate concentrations and meteorology. The spatial, seasonal patterns of light scattering coefficients to aerosol concentration were found to be readily explained in terms of their high degree of associations at most sites. Regression analyses relating light scattering coefficients to aerosol concentrations and relative humidity produced high levels of correlation and physically reasonable regression coefficients. This study has shown the key role played by particulate size in the atmospheric extinction. Those conditions that encourage the formation or growth of particles in the optically effective part of the accumulation mode are linked to reduced visibility. These results are in good agreement with Mie theory, and allow a much better match between theory and experiments than those gained by summing all mass below 2.5 μm or even key chemical constituents in the less than 2.5 μm regime.

It appears that each city had visibility-limiting aerosols which were unique and linked not only to the aerosol sources present, but also to topography and local meteorology. It should be noted that while Los Alamitos and Los Angeles had many visibility-limiting variables in common, each city also had site-specific influences of considerable importance to visibility. Therefore, the solutions for visibility problems should be based upon local studies which take into consideration the indigenous aerosols. Finally, this paper and some other studies (Macias *et al.,* 1981 and Trijonis *et al* ., 1982) have shown great sensitivity to particulate size and chemical species. It is perhaps instructive that many of these studies took place in the western United States, a region that undergoes dramatic changes in meteorology and consequently aerosol properties, even on a diurnal time scale. It would be unwise to use assumptions in the western United States that were developed for more stable conditions, especially since the scenic resources of the west often depend on visibilities almost never seen in urban areas of the eastern half of the country.

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