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SUMMARY OF CONCLUSIONS
AND RECOMMENDATIONS FROM A
VISIBILITY SCIENCE WORKSHOP

Technical Basis and Issues for a National
Assessment of Visibility Impairment

National Center for Atmospheric Research
Boulder, Colorado
September 19-20, 1991

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MASTER

Summary

A workshop was held in Boulder, Colorado, on September 19-20, 1991, at the National Center for Atmospheric Research to assist the U.S. Department of Energy, Office of Fossil Energy, in developing a national assessment of visibility impairment. Participants reviewed the current technical basis of the relationships between anthropogenic emissions of SO₂, NO_x, and volatile organic compounds and visibility impairment; reviewed the major scientific issues and uncertainties related to visibility-impairing aerosols; and recommended technical approaches for performing such an assessment.

Two documents were used in the workshop discussions: a scoping study report that provided a preliminary characterization of visual air quality changes, and a working paper that highlighted technical issues related to visibility and aerosols and proposed several approaches for the assessment, which will be produced in 1992.

The workshop panel made the following comments and recommendations with respect to the 1992 Assessment:

- The approach described in the scoping study is an appropriate starting point for characterizing visibility improvements that might be attainable by sulfate reductions in different areas of the country.
- There is clear evidence that aerosol light extinction efficiencies vary substantially with relative humidity and particle size distribution.
- Uncertainties in measured carbon aerosol loadings, both total mass and size-resolved mass concentrations, are poorly understood. There are major uncertainties for this aerosol species that frequently comprises a significant fraction of the aerosol mass and, therefore, can have an effect on atmospheric optics that is difficult to quantify.
- Review and evaluate available aerosol concentration data for use in driving the assessment. Use intensive measurement data from a variety of locations to augment the routine network data on diurnal variability and size-resolved chemical composition.
- Use visual range in addition to aerosol extinction coefficient as an indicator of visual air quality. Replace the "visual clarity classes" of the scoping study with frequency and/or probability distribution curves. Add a table relating the angular sizes of scene elements to contrast thresholds and visual range.

- Estimate uncertainties and variances in derived extinction coefficients from uncertainties and variances in scattering efficiencies and relative humidity response functions, and aerosol concentration and relative humidity data, respectively.
- Develop the design plan for the 1994 Assessment concurrently with the preparation of the 1992 Assessment. The plan should allow for improvements to the estimation of aerosol scattering efficiencies and relative humidity response functions derived from additional field measurements and include procedures for adapting existing aerosol chemical models for use in the analysis. Regional models are particularly important for relating emission changes to changes in secondary aerosol loading and, therefore, visibility responses.
- Include a section in the assessment that analyzes other effects of aerosols, such as climate forcing, when considering scenarios for improving visibility.

In developing a design plan for future assessments, the panel recommended the following questions for further study:

- What is the effect of various atmospheric processes on the size distribution of major chemical constituents, and what is the role of primary emissions, particularly elemental carbon, in controlling aerosol number size distributions and concentrations?
- How can aerosols be characterized with respect to size-resolved composition and mixing characteristics of major chemical species, including water, and how does this characterization relate to the optical properties of individual particles and integral optical properties of aerosol populations?
- How are mechanistic air quality models applied for addressing "what if?" questions and characterizing atmospheric conditions for various control options, where extrapolations from current conditions may be inappropriate?
- What defensible approaches are available or need to be developed to relate the quantitative measures of visual air quality (light extinction) to human perception indicators, such as distance, color, and contrast perception, of various outdoor scenes? How can incremental changes in aerosol mass loadings from ambient or background conditions be related to visual perceptibility?

Acknowledgments

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Introduction

The U.S. Department of Energy (DOE) has the responsibility for developing a National Energy Strategy (NES) to identify appropriate actions that will promote an adequate supply of energy for residents of the United States and, at the same time, enhance the environment and maintain national economic competitiveness in the world economy. Improvement of visibility has been identified as an important national goal and is, therefore, an important focus of the NES.

The linkage of visibility impairment trends with atmospheric aerosol sulfate trends has been reported in such recent studies as the National Acid Precipitation Assessment Program (NAPAP) State of Science/Technology Report (Trijonis et al. 1990) and the Electric Power Research Institute (EPRI) Analysis of Alternative SO₂ Reduction Strategies (EPRI 1991).

These reports showed general trends between visibility (visual range) reductions and sulfate increases and provided a representation of how the average annual visual air quality at selected locations might be altered by reductions in SO₂ emissions. Conclusions drawn from these representations, however, may have serious biases because several nonlinear processes were not considered, including nonlinearities in sulfate formation, aerosol particle growth in the presence of atmospheric water vapor, and aerosol optical phenomena attributable to non-homogeneous aerosol chemical mixtures. In particular, the depiction of the average visibility at various locations in the country may be biased because the calculations were based on average annual relative humidity (RH) values. Thus, these results do not account for the nonlinear response of hygroscopic aerosol light scattering to variable RH conditions in the atmosphere, which may show significant variations on time scales as short as 1 hour.

To evaluate the potential magnitude of such biases and to get a general indication of the kind of visibility improvements that might be attained by sulfate reductions in different regions of the country, a scoping study was performed.^(a) In this study, approximate seasonal sulfate air concentrations were adapted from NAPAP (Trijonis et al. 1990) and used as daily values. Four locations in the United States representing contrasting differences in sulfate concentrations and RH were selected for the study. Aerosol extinction coefficients were estimated from empirical relationships between particle light-scattering extinction and fine particle mass concentration. Hourly RH values at each location, derived from local National Oceanic and Atmospheric Administration climatological data for each location, were used to adjust the aerosol light extinction coefficients according to empirically derived RH response functions. To study the effect of sulfate reductions, ambient sulfate concentrations were changed according to the emission change scenarios of the 1990 NAPAP Assessment. The major finding of the scoping study was the sensitivity of the results to using highly time-resolved RH data.

(a) 1991 draft report, Office of Fossil Energy, U.S. Department of Energy, Washington, D.C.

The scoping study provided a relatively simple, but empirical, approach for developing an assessment of visibility impairment in the United States. A more credible assessment, however, requires a thorough examination of the relationships among precursor emissions, aerosol concentrations, aerosol light extinction, and RH effects. One such approach would be to couple a suitable air quality model with aerosol microphysical/chemical and optics modules and to use appropriate input data (meteorological variables, aerosol precursor emissions, and aerosol and trace gas concentrations) to generate the necessary annual visibility characterizations. It is evident, however, that some blend of empirical approaches and mechanistic modeling will be necessary to produce the improved periodic assessments in a timely manner.

Workshop Purpose

The Visibility Science Workshop was held in Boulder, Colorado, on September 19-20, 1991, at the facilities of the National Center for Atmospheric Research (NCAR). The purpose of the workshop was to review the current technical basis of the relationships between anthropogenic emissions of SO₂, NO_x, and volatile organic compounds (VOC) and visibility impairment. Participants also reviewed the major scientific issues and uncertainties related to visibility impairing aerosols that need to be addressed in the development of a national assessment of visibility impairment.

The workshop panel was instructed to identify specific areas where improved scientific understanding and technical information could be expected to reduce uncertainties in the assessments. Specific areas to be considered included empirical and theoretical relationships between aerosol light scattering extinction, size-resolved chemical aerosol species concentrations, relative humidity, and visual air quality perception.

Two working documents were the focus of the workshop discussions: 1) A scoping study report, which provided a preliminary characterization of changes in visual air quality when sulfate concentrations are changed from current conditions and when short time-scale variations of ambient relative humidity are considered, and 2) a working paper, which highlighted a number of technical issues related to visibility and aerosols and proposed several approaches for the assessment. The workshop panel was also asked to review the technical approaches presented in the working paper, identify strengths and weaknesses of those approaches, and recommend alternative approaches for use in performing an assessment, including the selection of appropriate aerosol mass concentration/extinction algorithms, relevant air quality and aerosol models, and suitable data sets for evaluating the algorithms/models.

Assessment Purpose and Client

The purpose of a national visibility impairment assessment in the present context is to provide DOE a technical basis for energy policy decisions related to the benefits of improved visibility that might be expected from emission reductions. A general energy policy change, for example, that would lead to emission reductions is a shift from the use of coal to natural gas and nuclear sources. The focus of DOE's interest in this assessment is to continue support for the development of the NES.

Assessment Structure, Plan, and Schedule

The major deliverable product of the project is a national assessment of visibility impairment. The first step in this activity is the development of a workplan that identifies the methods, tools, and data that will be needed to perform the assessment. In the development of this plan, the project will rely on the expert advice of the scientific community through two workshops: a Visibility Science workshop, as described in this report, and a Visibility Assessment Workshop. The latter will focus on 1) a review of the assessment workplan and its responsiveness to DOE policy and energy strategy needs; 2) the context of the DOE assessment with respect to the visibility assessment activities of NAPAP and other federal, state, and private agencies; and 3) the identification of additional research needs for accomplishing the assessment activities of DOE and other agencies with respect to scientific/technical issues, policy/strategy issues, and valuation issues.

The 1992 Assessment, which is currently in review, will use currently available methods, models, algorithms, and data. It will also estimate the uncertainties in the assessment results. After its completion, future assessments will be prepared biennially, as new knowledge, improved analysis methods and tools, and more data, derived from research and field measurement activities in related efforts within DOE or other agencies, are brought to bear on the problem.

The remainder of this report contains a review and evaluation of the scientific issues addressed in the workshop, including relationships among visual air quality, aerosol characteristics and environmental conditions; the application and selection of assessment models; relationships between visual air quality indices and aerosol optical properties; and the aerosol/climate-forcing problem and visibility. Approaches are described for both the 1992 Assessment and future assessments. A list of the workshop panel members is given in the Appendix.

Scientific Issues

The Visibility Science Workshop was organized around three major technical themes:

1. relationships among aerosol light-scattering (extinction) coefficient, aerosol chemical and physical characteristics, and environmental conditions, such as relative humidity and temperature
2. microphysical and aerosol optics models for calculating aerosol chemical mixing characteristics and optical properties, and their incorporation into regional-scale air quality models
3. characterization of visibility perception attributes or other indices of visual air quality in terms of light extinction coefficient.

In addition, a discussion of the overlap of data and information needs for visibility impairment studies and the emerging issue of aerosol/ climate radiative forcing, which is expected to have a prominent role in the U.S. Global Change Program, was included as a workshop topic.

A central focus of the workshop was to review and evaluate the scientific issues that need to be considered in characterizing the high time-resolution variability of visibility impairment nationally and estimating the anticipated changes in visibility impairment that would result from a reduction of the anthropogenic emissions of SO_2 , NO_x , and VOC and the attendant reductions in secondary aerosol particles (sulfate, nitrate, and organic compounds) formed in the atmosphere from these precursor gases.

A key conclusion of the workshop panel was the need to understand the variability of sulfate aerosol, which has been identified as an important component of visibility-reducing haze in many areas (except possibly for the mountainous West), and how sulfate covaries with relative humidity and other aerosol chemical components. These issues are discussed in the context of the major themes in the following sections.

Relationships Among Visual Air Quality, Aerosol Characteristics, and Environmental Conditions

The light extinction coefficient, b_{ext} , is the controlling physical variable in quantifying the effects of fine particles on visual air quality, solar irradiance at the ground, and the resulting climatic forcing. Furthermore, all indices of visual air quality ultimately reduce to the light extinction coefficient as the generally accepted objective, measurable variable. The extinction coefficient is a measure of the light attenuation as a result of scattering (b_{scat}) and absorption (b_{abs}) by gas molecules and aerosol particles along the direction of propagation of the light beam, as expressed in the Beer-Lambert law

$$dI/I = -b_{\text{ext}} dx = -(b_{\text{scat}} + b_{\text{abs}}) dx = -(b_{\text{RG}} + b_{\text{sp}} + b_{\text{ag}} + b_{\text{ap}}) dx \quad (1)$$

where I = intensity of the light beam

dI = the change of intensity as the light beam traverses a distance, dx

b_{RG} and b_{sp} = the molecular (Rayleigh) and particle scattering extinction coefficients, respectively, and

b_{ag} and b_{ap} = the molecular and particle absorption coefficients, respectively.

The scattering coefficient is usually much larger than the absorption coefficient.

Other quantities of interest to visual air quality or climate forcing that are related to the extinction coefficient include visual range, L_v , as defined by the Koschmieder relationship,

$$L_v = \ln \epsilon / b_{ext} = 3.9 / b_{ext} \quad (2)$$

and the optical depth, δ , expressed by

$$\delta = - \ln (I_{trans}/I_0) = \int_0^{\infty} b_{ext} dz. \quad (3)$$

Here ϵ is the contrast threshold for a large black object viewed against the horizon sky during daytime with uniform illumination. The optical depth, which is the integral of the extinction coefficient over altitude z , is a measure of the total attenuation of the direct solar beam at the ground.

Aerosol light extinction coefficients can be measured directly or can be calculated by applying Mie theory to measurements of size-resolved chemical composition (see, for example, Waggoner et al. 1981, Sloane 1986, White 1986, and Zhang 1990). It is observed that the aerosol light-scattering coefficient is directly proportional to total fine particle mass, M , as expressed by

$$b_{sp} = \alpha \cdot M \quad (4)$$

where α is the scattering efficiency. The reason that fine particles ($<2 \mu\text{m}$ diameter) provide the dominant contribution to light extinction is that the single particle optical cross section (as calculated from Mie theory) per unit particle volume reaches a maximum for particles with diameters around $0.5 \mu\text{m}$, as shown in Figure 1. Typical values of α lie in the range of 3 to $7 \text{ m}^2/\text{g}$. Although the cause of the variability in α is understood as a function of particle size and chemical composition through Mie theory, it is difficult in any given situation to calculate α precisely. This uncertainty arises because the chemical (mixing characteristics) and physical (size distribution) variability of aerosol populations is often poorly determined. Typically, under clean conditions, the visual effects of aerosols are determined by only a relatively small quantity of fine particle aerosol mass—about $1 \mu\text{g}/\text{m}^3$.

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Figure 1. Particle Extinction, Scattering, and Absorption Cross Sections Per Unit of Aerosol Volume, as Calculated from Mie Theory (from Charlson et al. 1984).

Aerosol light extinction is further modified by the relative humidity response factor, $f(RH)$, to account for the effect of atmospheric water vapor on particle growth and increased aerosol scattering efficiency. The RH effect has been demonstrated both experimentally and theoretically. The humidity response function can be determined in one of three ways: 1) aerosol samples are weighed as a function of RH and extinction is calculated using Mie theory and an assumed size distribution, 2) it is measured directly, or 3) Mie theory is applied to measured size distributions and chemical composition as a function of RH.

Method 1 is the classical approach begun in the 1960s. Method 2 was started by Covert in the early 1970s (Covert et al. 1980; Waggoner et al. 1981). In method 2, data are typically presented as plots of the ratio of measured light scattering coefficient to that measured at a reference RH as a function of RH. Composite behavior is shown in Figure 2. While there are frequently many similarities between the curves, there is no one humidity correction function

Figure 2. Measured Enhancement of Scattering as a Function of Relative Humidity. The hatched area contains the complete range of ambient measurements (from Covert et al. 1980).

that can be used everywhere, because $f(RH)$ is very dependent on chemical and microphysical variables. Method 3 is a detailed microphysical approach being developed and refined by McMurry and colleagues (Zhang et al. 1991). Examples of sulfur light-scattering efficiencies as a function of RH and mass median diameter from measurements at the Grand Canyon during the 1990 NGS Visibility study are shown in Figure 3.

Finally, aerosol light extinction can also be measured at ambient RH conditions, but these observations are difficult to interpret and extrapolate to other meteorological conditions without an independent determination of the RH response function. Hence, the preferred observational mode is to measure aerosol light scattering at a reference dry condition and apply the appropriate response factor.

Multiple regression analyses of observations of aerosol fine particle mass composition have been used to derive empirical relationships between aerosol light-scattering extinction coefficient and the mass concentration of individual chemical components of aerosol populations. One form is expressed as linear combination of species mass concentrations:

$$b_{sp} = \sum_i \alpha_i \cdot f_i(RH) \cdot M_i \quad (5)$$

where M_i = the mass concentration of aerosol species i

α_i = the fitted scattering efficiency for species i at reference relative humidity RH_0 (typically at some low value between 3% - 30%),

$f_i(RH)$ = relative humidity response factor.

Figure 3. Calculated Ammonium Sulfate Light-Scattering Efficiencies for Aerosols at the Grand Canyon as a Function of Relative Humidity. From a) low-pressure impactor sulfur size distributions and a selected, averaged tandem differential mobility analyzer (TDMA) size, and b) as a function of particle sulfur mass-median diameter (from Zhang et al. 1991).

Equation (5) can also be derived from Mie calculations, but only if the different chemical components of the aerosol population are independent of one another (externally mixed). Generally, most ambient aerosol particles are chemically inhomogeneous (internally mixed). In such cases, b_{sp} cannot be expressed as a linear combination of M_i (Sloane 1986; White 1986). Nevertheless, this form is frequently used as an approximation for attributing visibility impairment to various aerosol chemical species.

Chemical reactions induced by sunlight between a variety of anthropogenic and natural precursor gas emissions (primarily SO_2 , NO_x , and VOC) are responsible for producing most of the optically active fine-particle aerosols (sulfate, nitrate, and organic particles) in the atmosphere. Generally, these chemical and other physical processes lead to chemically heterogeneous aerosol particle mixtures, whose optical effects can be very different.

Atmospheric aerosols also originate as primary (direct) emissions from combustion processes (e.g., elemental carbon, sulfate, fly ash) and mechanical processes (e.g., dust, sea spray). The latter type of aerosols, especially elemental carbon, may act as seeds for sulfate or organic vapor condensation, although this is largely a hypothesis at the present time. It is often observed that aerosols containing sulfate are the dominant chemical component influencing b_{sp} , especially in the eastern half of the United States, but it is also observed that the sulfate fraction of aerosol mass is highly variable and that the coupling (covariability) between different aerosol chemical fractions is difficult to characterize. Moreover, in areas where carbon aerosol is important, the relative partitioning between the organic and elemental forms, which in itself is a difficult measurement to make, depends on knowledge of the many organic carbon sources, such as terpenes, anthropogenic hydrocarbons, and biomass combustion products, which are also not available.

In spite of these difficulties, the characterization of aerosol chemical variability is improving as a result of data gathered during recent intensive field measurement studies. Some examples follow.

1. Studies performed at the Grand Canyon (Zhang et al. 1991) showed that aerosol size distributions varied significantly from day to day and that this observed variability appeared to be related to gas-phase sulfate production mechanisms, which are dependent on relative humidity, because the size distributions, sulfate mass, and relative humidity showed a strong covariance. There was also evidence that elemental carbon particles acted as seeds for sulfate condensation, because sulfate and carbon were often found associated in the same particles. Another hypothesis is that the sulfate/carbon aerosol acted as cloud condensation nuclei (CCN), with additional sulfate being produced via aqueous-phase processes. References that further discuss the relationship between size distributions of secondary sulfates and the chemical mechanism of sulfate formation include Hering and Friedlander (1982), McMurry and Wilson (1983), and John et al. (1990). The observations discussed in these journal articles will play a major role in establishing a quantitative and mechanistic link between atmospheric transformations for sulfates and visibility impairment.

2. Recent measurements suggest that the major aerosol chemical species (typically organic and elemental carbon and sulfates), including those found in Los Angeles and the Grand Canyon, are to some extent externally mixed. Mixing characteristics affect light-scattering efficiencies as well as chemical transformations that occur within particles (McMurry and Stolzenburg 1989; Hering and McMurry 1991; Zhang et al. 1991).
3. An interaction between sulfate, nitrate, and ammonia was apparently observed during the Denver Brown Cloud study, where aerosol nitrate appeared to compensate for a decrease in sulfate (SO_2 was reduced in Denver during the study by switching power plants from coal to gas), resulting in no net effect on visibility or light extinction (Watson et al. 1988). This interaction is hypothesized to occur because more ammonia, which would normally be associated with the sulfate, would be available to react with gas-phase nitric acid. The Denver study may be exceptional, because in many areas of the country, ammonia is very limited, and major sulfur reductions would be needed for this mechanism to become significant. Moreover, ammonium nitrate is rather volatile and may be mostly in the gas phase, depending on temperature and relative humidity. Finally, it should be pointed out that other SO_2 sources outside of Denver were not modified and may have influenced the results of the study.

Interactions between different aerosol fractions are often not well characterized with respect to chemical composition and size. An understanding of these interactions is crucial when dealing with questions about the incremental impact on extinction (visibility) of removing individual chemical components (e.g., emissions controls) in the context of contributions from all other remaining species. Reducing sulfate, for example, by a factor of two might not necessarily reduce aerosol light extinction by the same factor, because of the interactions of the remaining components and the manner by which sulfate is distributed as a function of particle size before and after its removal. Furthermore, because of different responses to relative humidity changes, the effect of removing hygroscopic aerosol such as sulfate and nitrate on the relative change in the water content of the aerosol at a given ambient relative humidity will be greater than that of removing a less hygroscopic or hydrophilic aerosol from the population.

Finally, if the aerosol particles consist of both water-soluble and insoluble components, such as in the Grand Canyon sulfate/carbon aerosol example given above, then removing sulfur and/or carbon (as a potential means of reducing the visibility impairment because of these aerosols) requires knowledge of how these chemical components are to be removed from the aerosol population and from what size ranges. For example, if carbon particles act as seeds for sulfate condensation, then changing the number of carbon seeds could possibly affect the sulfur size distribution; i.e., a smaller number of seed particles might lead to a larger average size for sulfate particles. Much work remains, however, to sort out the chemical and physical processes and interactions that occur in the atmosphere.

The conclusion that can be drawn from the current understanding of chemical variability and heterogeneity of mixing characteristics of aerosol populations is that the relevant details of the aerosol microphysical structure (internal versus external mixing, size dependence of composition, and hygroscopic and optical properties as a function of humidity) cannot be used directly in atmospheric optical calculations. Instead, the effect of processes may be better understood in the context of an air mass history, as simulated, for example, in a large-scale

atmospheric chemical transport/air quality model that includes the relevant formulation of these microphysical/chemical processes.

There are also pronounced regional differences in aerosol chemical characteristics in the United States that must be considered in the visibility assessment. In the eastern part of the country, for example, sulfate aerosol is the dominant chemical form affecting visibility, while in the West (e.g., in the Grand Canyon), carbon and coarse (soil) particles contribute more to light scattering than sulfate. The interaction between sulfates and other species, as mentioned above, also appears to have important geographic differences. Currently available aerosol chemical data are still incomplete for the country as a whole, but data on some aspects of aerosol chemical and optical properties are becoming available from routine monitoring studies, such as the National Park Service (NPS) IMPROVE network, and from intensive measurement programs at a variety of sites, including Denver (1982 and 1987), Los Angeles (1987), Phoenix (1989-90), Detroit (1981), Uniontown (1990), Salt River Project/Grand Canyon National Park (1990), and Shenandoah National Park (1991), where detailed size-resolved aerosol chemical information was collected.

The covariance of aerosol concentrations, particularly sulfate, and environmental conditions, such as relative humidity, is also important in characterizing visibility and its temporal variability. These quantities often covary in different ways in different parts of the country and can lead to pronounced effects on light extinction. In the scoping paper, the sulfate concentration was assumed constant throughout a season, while actual sulfate levels showed a great deal of diurnal and day-to-day variability. For example, airborne sulfate, which tends to vary episodically, might be found to predominate one day out of every three at some location. In this example, if sulfate were reduced by some factor, an improvement in visibility might also be seen for only one day out of every three, but, then again, the effect might not be very pronounced, depending on the other aerosol present and the environmental conditions.

To summarize, there are many factors that need to be known and understood to estimate light extinction by aerosols for use in a national assessment of visibility impairment. These include

1. emission rates of gases that are precursors to secondary aerosols such as SO_2
2. aerosol formation mechanisms and rates
3. aerosol residence times
4. combining items 1-3 or from independent measurements, aerosol mass concentrations as a function of chemical composition
5. aerosol light-scattering efficiency as a function of chemical composition
6. the aerosol light-scattering humidity response function.

Some of these factors are better understood than others. Estimates of the variability and uncertainty in these factors are given in Table 1. When all of these estimates are combined, the overall uncertainty in the calculation of light extinction from all sources of variability and uncertainties is on the order of a factor of two.

Table 1. Factors Contributing to the Variability and Uncertainty in Aerosol Extinction Coefficient (adapted from Charlson et al. 1992)

Quantity	Value	Relative Variability or Uncertainty (%)
Emission Rate Q_{SO_4} (Anthropogenic)	70 Tg/a	15
Q_S (Natural)	35 Tg/a	20
Conversion Fraction Y_{SO_4}	0.5	50
Residence Time τ_{SO_4}	0.02 a	50
Extinction Efficiency α_{SO_4}	5 m ² /g	40
Humidity Response Function $SO_4 f(RH)$ at RH = 0.8	1.7	20
Hemispheric Backscatter Fraction	0.15	30

There are three experimental approaches for determining aerosol light extinction: 1) measure aerosol light-scattering coefficients and aerosol dry mass, 2) use multivariate analysis methods to apportion extinction factors to each chemical component, and 3) apply Mie theory to measurements of size-resolved chemical composition and, if available, mixing characteristics and water content of aerosol populations. It is expected that these three approaches should converge, because theoretically, the relationship between light extinction and aerosol size and chemical composition is well understood. At the present time they do not.

Panel Recommendations

To bring closure among and validate the various methods for determining aerosol extinction, the workshop panel made the following comments and recommendations.

1. An integrated series of measurements must be performed to relate size-resolved composition, mixing characteristics, and physical properties of the major chemical aerosol species, including water, to aerosol optical properties (scattering extinction and angular scattering extinction coefficients) through the Mie formalism and through comparison to observations. Such an integrated set of measurements is needed for individual particles and aerosol particle populations.

2. Additional data are required to validate the theoretical/model predictions and empirical relationships for aerosol light extinction as a function of RH and aerosol composition and size, particularly for the dominant forms of ambient sulfate (H_2SO_4 , NH_4HSO_4 , and $(\text{NH}_4)_2\text{SO}_4$) and for other important species (organic and elemental carbon, nitrate, soil dust, etc.). Data are also needed to understand how water is bound up in the aerosol and if it is bound preferentially to particular species of particles. There is also a need to understand how carbon is split between hygroscopic and nonhygroscopic particles.
3. Numerical studies to examine the incremental impact of each species on light extinction are useful exercises. In such studies, scattering efficiencies are assigned to various fractions of aerosol chemical species and then the relative concentrations of these component species are varied, while keeping other factors the same. Caution must be exercised, however, because the mixing characteristics of typical aerosol populations are much too complex to sort out the effects of atmospheric chemical and physical processes and the distribution of the different chemical components as a function of size.
4. Further study is needed to test hypotheses of how various atmospheric chemical and physical processes affect the distribution of major chemical constituents (sulfate, nitrate, organic and elemental carbon species, water, dust, sea salt) as a function of size. There is also a need for better understanding of the role of primary emissions of aerosol species, particularly elemental carbon, in governing the size distributions of secondary sulfates, either as nuclei for sulfate condensation or as sites for chemical reactions.
5. Uncertainties in measured carbon aerosol loadings, both total mass concentrations and size-resolved mass concentrations, are poorly understood. Discrepancies between carbon data obtained with different samplers and analyzed with different methods can produce significantly different results, especially in pristine areas where aerosol mass concentrations are low. Visibility impairment is often an important issue in such locations. There are major uncertainties for this aerosol species, which frequently comprises a significant fraction of the aerosol mass and, therefore, can have a large effect on atmospheric optics that is difficult to quantify.
6. Aerosol chemical measurements can only be made at single points in space; this requires that the aerosol light-scattering extinction must also be measured at the same points. The role of spatial inhomogeneities can be evaluated by using the observed temporal variability in relationship to the observed meteorological conditions. Measurements are needed at selected locations to learn about the fundamental processes that are occurring and to get practical knowledge for visibility- and climate-related programs. They are needed at both clean locations (e.g., Cheeka Peak, Washington, and near Laramie, Wyoming), as well as at impacted ones (e.g., Los Angeles, Denver, St. Louis and surroundings, New York City, Norfolk, Shenandoah National Park). Data from clean locations are needed to understand departures from pristine situations. These data should also be linked to aerosol measurements at remote climate monitoring stations, such as at Sable Island (Nova Scotia), Cape Grim (Tasmania), and Bermuda.

7. Real-time measurements of aerosol chemical species are difficult to make, but some progress has been made with respect to real-time measurement capability for sulfate (Garber et al. 1983; Cahill et al. 1987) and organic and elemental carbon (Turpin et al. 1990). Newer and smarter versions of older instruments, such as the integrating nephelometer and the light-scattering humidograph, are needed for application in the next generation of field studies.

With respect to estimating aerosol light extinction for the 1992 Assessment, the workshop panel made the following comments and recommendations:

1. The approach described in the scoping study is an appropriate starting point for characterizing visibility improvements that might be attainable by sulfate reductions in different areas of the country.
2. There is clear evidence that aerosol light extinction efficiencies vary substantially with relative humidity and particle size distribution. Multiple linear regression produces values for species-specific aerosol scattering efficiencies appear to be accurate to within a factor of approximately two. While these efficiencies may be used in the 1992 Assessment, they need to be reviewed and updated using available experimental data. A literature review would reveal biases, as well as identify available measurements, especially those that have been made in urban locations.
3. Available aerosol concentration data, such as those from the IMPROVE network, need to be reviewed and evaluated for use in driving the assessment over an annual cycle. Day-to-day variations can be derived from selected intensive study data bases, because they may be the best available source in terms of frequency and quality. Attention must also be paid to selecting appropriate climatological data. RADM simulations, which were performed for the 1990 NAPAP Assessment should be examined for augmenting the aerosol concentration and climatological data, particularly for characterizing diurnal cycles and day-to-day variations.
4. Uncertainties and variances in derived extinction coefficients and visual ranges need to be estimated from uncertainties and variances in scattering efficiencies and relative humidity response functions, and aerosol concentration and relative humidity data, respectively. The results should be folded into a simple algorithm to show the sensitivity of the conclusions to these uncertainties and variances.

Application and Selection of Models for Assessments

Measurements of atmospheric aerosol properties and humidity are often by necessity made at one or a few points in space and over a limited duration in time. So to generalize over larger spatial and temporal scales, it is necessary to resort to the use of models. The models must be integrative in the sense of extending from the source region of emitted gases, such as SO₂, which are precursors to secondary aerosols, to the consequent optical impact (visual air quality) region. Data are required under an adequately variable set of conditions to test the models and define their uncertainties.

Mechanistic models attempt to incorporate mathematical descriptions of the important chemical and physical processes needed to investigate source-receptor relationships and other atmospheric phenomena of interest. Observational data are used mainly to establish initial and boundary condition estimates and to evaluate model performance. Empirical models, which tend to make more extensive use of observations rather than fundamental process descriptions, are also used to establish source-receptor relationships. Generally relationships deduced from empirical models are valid only for the time and location for which the observations were taken. For this reason, the more comprehensive deterministic models provide the most appropriate and most detailed means of assessing the impacts of projected emission changes on source-receptor relationships and future visibility changes. Mechanistic models, therefore, are appropriate tools to be used in combination with any empirical approaches developed for the assessment.

The application of the more complex mechanistic models is particularly useful for examining the combined impacts of sulfur, nitrogen, and carbon emissions on visibility impairment over a variety of regions that are influenced by different atmospheric and emission conditions. They are also invaluable for performing exercises that cannot be done experimentally.

The accuracy of current models is limited by an incomplete understanding of the various atmospheric processes, including aerosol phenomena, such as aerosol size distribution and chemical mixing characteristics. Evaluation of the models require data bases that have been acquired for the same time periods and locations for which the simulations will be performed.

A hierarchy of models are available for application to atmospheric and visibility studies. The most complex are the regional air quality modeling systems; the components within such a hierarchy are illustrated schematically in Figure 4. These modeling systems contain micro-physical and chemical modules that account for aerosol mixing characteristics when several chemical components are present. The evolution of a series of these models is shown in Table 2. Other models, such as chemical mass balance receptor and single-source plume types, which use various secondary aerosol models or empirically determined data on aerosol properties, are summarized in Table 3.

A version of the Regional Acid Deposition Model (RADM), which was originally developed for NAPAP (Chang et al. 1987; 1990), has been applied to examine regional sulfate patterns (Middleton et al. 1991; Trijonis et al. 1990) has also been adapted for and is being used in the Denver Brown Cloud II study (Middleton and Burns 1991). In the Brown Cloud study, the model, the Denver Air Quality Model (DAQM), includes various secondary aerosol processes and aerosol optics parameters for visibility calculations (Figure 4). DAQM currently uses a modified version of MARS for inorganic species (Saxena et al. 1986; see also Table 2) and the organic gas-to-particle conversion scheme of Grosjean and Seinfeld (1989). Empirical extinction efficiencies for sulfate, nitrate, and elemental and organic carbon particles are currently used to calculate extinction coefficients and other aerosol optical parameters from concentration fields calculated by the model (Table 4). The effects of water (RH) are also accounted for by empirical data, as shown in Figure 5.

Mechanistic Modeling System

Figure 4. Mechanistic Modeling Framework. Major development efforts are required for the aerosol and visibility assessment components highlighted in the shaded boxes.

Table 2. History of Aerosol Models

Model Name and Type	Gas Phase	Aerosol Phase	Source
NH ₄ NO ₃ equilibrium	NH ₃ and HNO ₃	NH ₄ NO ₃	Stelson and Seinfeld (1982); Russell et al. (1983)
EQUIL: Sulfate and nitrate equilibrium	NH ₃ and HNO ₃	NH ₄ NO ₃ , ammonium sulfates and mixed salts	Bassett and Seinfeld (1983)
MARS: Fast sulfate and nitrate equilibrium	NH ₃ and HNO ₃	NH ₄ NO ₃ , ammonium sulfates	Saxena et al. (1986)
EQUILIB: NaCl added to MARS	NH ₃ , HNO ₃ , and HCl	Numerous salts of ammonium, sodium, nitrate, chloride, and sulfate	Pilinis et al. (1987)
SEUILIB: Expand EQUILIB to calculate size distribution	NH ₃ , HNO ₃ , and HC	Size-distributed ammonium, sodium, nitrate, chloride, and sulfate	Pilinis and Seinfeld (1987)
AIM: Transport considerations taken into account	NH ₃ , HNO ₃ , and HCl	Size-distributed ammonium, sodium, nitrate, chloride, and sulfate	Wexler and Seinfeld (1991)
SOAM: Secondary organics added to AIM	NH ₃ , HNO ₃ , HCl, and condensable organics.	Condensable organics and inorganics	Pandis et al. ^(a)

(a) Submitted to Atmospheric Environment.

Specifying emissions accurately for models of this type is difficult. There are still large uncertainties in nitrogen and carbon emissions in comparison to sulfur. Such uncertainties become especially important when attempting to depict secondary aerosol formation accurately.

Table 3. Visibility Source Apportionment Methods

Regional Haze Assessment

- Speciated Rollback Models
- Combinations of chemical mass balance receptor models with various secondary aerosol models
- Mechanistic transport and secondary aerosol formation models with empirically determined aerosol size distribution data to facilitate light extinction calculations

Existing Single-Source Plume Analysis

- Simple photographic identification methods
- Combinations of chemical mass balance or tracer techniques with a credible secondary aerosol model
- Advanced reactive plume or plume blight models with empirically determined data on aerosol properties

New Single-Source Plume Analyses

- Advanced reactive plume or plume blight models with empirically determined data on aerosol properties

Table 4. Average Natural Background Extinction Efficiencies
(adapted from Trijonis et al. 1990)

<u>Fine Particles</u>	<u>Extinction Efficiency (m²/g)</u>
Sulfate	2.5
Nitrate	2.5
Ammonium	2.5
Water	5.0
Organic Carbon	3.75
Elemental Carbon	10.5
Dust	1.25
<u>Coarse Particles</u>	
Organic Carbon	0.6
Elemental Carbon	0.6
Dust	0.6

Figure 5. Empirical Scattering and Absorption Efficiencies for Various Aerosol Chemical Compositions as a Function of Relative Humidity

Panel Recommendations

With respect to the application of models for visibility assessment, the workshop panel made the following observations and recommendations:

1. Mechanistic air quality models, such as RADM, have an important role in addressing "what if?" questions and characterizing atmospheric conditions for various emission control options, particularly when extrapolations from current conditions using empirical models may be inappropriate.
2. Aerosol microphysical models, such as MARS or SEQUILIB, which account for aerosol chemical mixing characteristics when several chemical components are present, need to be evaluated against available data for use in the assessments.
3. Evaluation of the models require data bases that have been acquired for the same time periods and locations for which the simulations will be performed. Moreover, field measurement efforts focused on integrated measurements of aerosol physical, chemical, and optical properties need to be closely coordinated with the development and use of the models to insure that the best possible tools are available for the assessment in a timely fashion.

4. Because resource limitations in turn limit the direct application of complex mechanistic models in some situations, approaches that use the simpler, more empirical models in combination with the mechanistic models should provide sufficient detail and predictive accuracy for use in visibility assessment applications. Such approaches need to be identified, tested, and evaluated.
5. The design plan for the 1994 Assessment should be developed concurrently with the preparation of the 1992 Assessment. The plan should allow for improvements to aerosol scattering efficiencies and relative humidity response functions derived from additional new field measurements and include procedures for adapting existing aerosol chemical models, such as MARS and SEQUILIB, and regional air quality models (with appropriate aerosol and optics modules), such as RADM, for use in the assessment analysis. The regional models are particularly important for relating emission changes to changes in secondary aerosol loading and, therefore, visibility responses.
6. The models should be used to guide the assessment analysis, especially in dealing with questions of temporal (diurnal and day-to-day) and spatial (region-to-region) variability and in cases where data are either not available or lack sufficient resolution. RADM simulations, which were performed for the 1990 NAPAP Assessment should be examined for augmenting the aerosol concentration and climatological data, particularly for characterizing diurnal cycles and day-to-day variations.
7. Uncertainties in model calculations should be identified and quantified, especially with respect to emissions and aerosol parameters.

Relationships Between Visual Air Quality Indices and Aerosol Optical Properties

Atmospheric aerosol light extinction coefficient is generally accepted as the primary objective measurable visual air quality variable. It is what models can produce as well. Other indices of visual air quality may also be appropriate for presentation purposes in an assessment of visibility impairment. Ideally a suitable index should 1) be a derivable function of measurable atmospheric properties, such as the light extinction coefficient; 2) reflect regional differences in the physical, chemical, and optical aspects of visibility impairment; 3) reflect regional differences in human responses to changes in visibility; and 4) be understandable to policy makers and the public.

Any viable visibility index must consider observable atmospheric optical phenomena in the context of the classical human physiological responses (contrast threshold, threshold luminance, and visual acuity) so as to optimize their organization for application to problems of human perception.

Visual range, as defined in the Koschmieder relationship, has been commonly used as an indicator of air quality. The concept of visual range, however, is an abstraction that cannot be objectively measured. Moreover, it can be misleading, because the layperson may interpret it as the distance he or she can actually see real objects. Other indices have been proposed, such as the JNC index being evaluated by the National Park Service and U.S. Environmental

Protection Agency (Trijonis et al. 1990) or the VAQ index described by Middleton et al. (1984). The latter index, based on human judgments of visual air quality, was found to be linearly related to $\ln(b_{\text{scat}})$, as shown in Figure 6.

The following points highlight the current state of knowledge about visibility:

1. Perception of color and contrast form the basic connection between human perception and atmospheric properties.
2. Perception of contrast detail in the atmosphere is known to depend on size and context.
3. Theories of contrast perception based on laboratory studies are probably adequate for predicting thresholds and visual range of contrast detail as a function of size.

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Figure 6. Relationship Between Average Overall Visual Air Quality Index, as Perceived By a Group of Observers, and the Natural Logarithm of the Aerosol Light-Scattering Coefficient During the Winter 1982 Denver Brown Cloud Study

4. Theories of just-noticeable differences (or changes, JNCs) have not been validated for use in the atmosphere. They are, however, the only means to estimate changes in atmospheric clarity for complex scenes at super-threshold levels.
5. Perception of color in the natural atmosphere is not as well understood as perception of contrast.
6. Experiments in the natural atmosphere show that the most often used color index, delta E, is not a good predictor of color difference as observed in the atmosphere. Currently, there is not a measure of color that is applicable to atmospheric visibility.
7. Ordinary or computer-generated photographs are fairly successful in displaying contextual effects on visibility. However, photos distort the observed saturation of colors, especially greens, yellows, and reds.
8. Relationships have been found between psychological interpretation indices, such as perceived visual air quality index (PVAQI) or scenic beauty index, and contrast or extinction coefficient. However, these relationships are highly scene-dependent in ways that are not understood.
9. Fundamental objections to psychological indices have been raised on the grounds that the average person does not have an inherent opinion of the "visual air quality" that can be discovered by experiment.

Panel Recommendations

While there are still many problems in trying to relate perceptions to physical measurements, and much work remains to be done in this area, the workshop panel made the following recommendations with respect to the presentation of the assessment results in terms of visual air quality indices or indicators **in addition** to light extinction coefficients:

1. Visual range may be used as an indicator of visual air quality in addition to aerosol extinction coefficient. The "visual clarity classes" of the scoping study should be replaced with frequency and/or probability distribution curves. Furthermore, if visual range is used as an indicator, a table should be added that relates the angular sizes of scene elements to contrast thresholds and visual range.
2. Actual or simulated photographs for various lighting and aerosol loading conditions should be used to augment the results of the visual range frequency curves.
3. Other defensible approaches that are currently available or being developed for relating the quantitative measures of visual air quality (light extinction) to human perception indicators, such as distance, color and contrast perception, of various outdoor scenes need to be reviewed and evaluated.
4. Additional studies need to be conducted to relate incremental changes in aerosol mass loadings from ambient or background conditions to visual perceptibility.

5. Other studies could be conducted to

- define a simple, generic scene that contains contrast detail, a single luminance edge, and colored patches that can be used in radiative transfer model
- improve understanding of perception of contrast and color in the natural atmosphere through field studies that combine physical measurements and contrast and color-matching studies by onsite observers
- improve simulated displays of the results of air quality and visibility models using the improved understanding developed by the field studies recommended above
- use the improved simulated displays to study in a controlled manner the psychological and economic effects of changes in atmospheric properties and concentration.

Aerosol/Climate Forcing Problem and Visibility

The emerging issue of aerosol radiative forcing of climate and visibility impairment share the same need for information on the relationships between aerosol optical properties and size-resolved aerosol chemical mixing characteristics (Charlson et al. 1991, 1992; Waggoner et al. 1981). Fine particle aerosols can affect climate in two ways: 1) they reflect light back into space and 2) they act as condensation nuclei for forming clouds, which, in turn, may modify the reflectivity of cloud layers. Sulfate aerosols appear to be linked to both types of climatic effects.

Recent evidence suggests that the effects of aerosols on climate are comparable to the hypothesized greenhouse warming that would result from a doubling of CO₂, but aerosols lead to cooling (Charlson et al. 1990). This suggestion is based on the analysis of two bodies of information: 1) natural emissions of sulfur species from the oceans result in a significant mass of sulfate aerosol in the remote background atmosphere, and 2) anthropogenic emissions of sulfur have increased dramatically during the past century and have surpassed the natural emission rate (ca. 1960 for the global anthropogenic flux of sulfur; ca. 1930 for the northern hemisphere flux). Charlson et al. (1990, 1991, 1992) assert that the backscatter of solar radiation by non-cloud, anthropogenic sulfate aerosol particles in cloud-free air significantly reduces the incoming solar radiation over much of the northern hemisphere. The aerosol cooling effect, however, does not compensate for the greenhouse warming effect, because both have different meteorological effects and cannot be averaged over the planet. Moreover, the influence of the greenhouse effect is strongest in winter and the effect of aerosols is strongest in summer.

The climate issue, in addition to issues such as acid precipitation and regional oxidant formation, which have already been the focus of study during the past decade, are clearly important ones for energy decisions. There is the opportunity, however, to consider the common scientific problems associated with all of these issues together and to coordinate research efforts that show the common links between them and provide multiple benefits to the various assessment activities.

Panel Recommendation

In view of its potential importance to energy policy issues, the workshop panel made the following recommendation:

The assessment should have a section that takes account of other effects of aerosols, such as climate forcing, regional oxidant formation, and acid precipitation, when considering scenarios for improving visibility.

Assessment Approaches

The workshop panel reviewed several draft plans for producing a national assessment of visibility impairment and recommended appropriate technical approaches consistent with the phasing of the assessments described previously. These recommendations are consolidated in this section.

The technical approach consists of the following elements:

1. identification of the appropriate analysis tools for determining the relationships among
 - emissions of primary aerosols and gas species that are precursors to secondary aerosol formation and the resultant ambient aerosol mass loadings
 - size-resolved mass concentrations of the important aerosol chemical species (including the dominant sulfate forms) and aerosol light extinction coefficient
 - aerosol particle chemical and water content as a function of relative humidity
 - aerosol light extinction coefficient and other indices of visual air quality that may be useful for the presentation of assessment results to policy and decision makers
2. a procedure for how these tools will be evaluated and applied to produce the assessment
3. data sets of aerosol optical properties (e.g., aerosol light-scattering observations) that can be used to evaluate the appropriateness of the relationships
4. data sets that can be used to drive the assessment, such as the meteorological variables (relative humidity, temperature, occurrence of precipitation and fog) and aerosol and gas concentrations with appropriate temporal information (e.g., mean seasonal values and standard deviations, diurnal cycles)
5. areas of the country and representative sites for which the assessment will be performed
6. methods of computing and presenting the results for each site (statistics, graphs, and photographs) with respect to the derived visual air quality parameters or indices.

A comprehensive assessment process is shown schematically in Figure 7. The main focus of this work is highlighted in the boxes and circle.

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Figure 7. Schematic Overview of the Analysis Components in the Visibility Assessment Process

1992 Assessment

The 1992 Assessment is similar to and expands on the Scoping Study.^(a) It essentially uses the same set of empirical relationships between aerosol mass concentration and aerosol light-scattering coefficient used in the Scoping Study, but uses actual time-series data of aerosol mass concentrations and relative humidity from a few selected sites rather than average monthly or seasonal values. To establish diurnal cycles in these quantities and to provide additional verification of the empirical relationships, data from selected intensive periods are also used. The elements of the 1992 Assessment follow.

1. Tools/Relationships

- a) M_{aer} vs emissions: Vary only secondary particle mass (i.e., assume linear relationship between emissions and fine particles). Use selected VAQM (RADM with modified MARS aerosol and visibility optics modules) simulations to begin characterizing the limits of this assumption.

^(a) 1991 draft report, Office of Fossil Energy, U.S. Department of Energy, Washington, D.C.

- b) b_{ext} vs M_{aer} : Extinction efficiencies from Trijonis et al. (1990), with consistency checks with the Sloane procedure (Sloane 1986).
- c) b_{ext} vs RH and composition: Trijonis et al. (1990) consistency checks using MARS or SEQUILIB models in RADM.
- d) VAQ Index vs b_{ext} : Use visual range $L_v = 3.912/b_{\text{ext}}$ and deciview $dV = 10 \log (390/L_v)$

2. Review literature to select best set of parameters for extinction efficiencies and humidity response function. Calculate the aerosol light-scattering coefficient (b_{sp}) from particle composition and water content and compare to available nephelometer-measured values of b_{sp} . Review results of evaluation of MARS and SEQUILIB models with high-temporal-resolution data from various intensive field studies (e.g., Shenandoah National Park-summer 1991, Uniontown-summer 1990, Hopi Point-winter 1990) (see below). Initiate checking of VAQM and its associated aerosol optics models with same data set or Denver Brown Cloud data set.

Use empirical procedures to estimate hourly b_{sp} values for an entire annual cycle (current conditions or base case) from available aerosol mass concentration data with adjustments for RH and T based on seasonal and diurnal climatological information (see below). Estimate diurnal variations in aerosol mass concentrations from recent intensive field studies or other data sources, including RADM runs for the NAPAP 1990 Assessment (Chang et al. 1990) and factor these variations into the calculations. Repeat the calculations using dry aerosol mass concentrations that are scaled by factors of 1.2, 0.6, and 0.2, respectively. Characterize limits of linear sulfate response assumption with selected simulations with VAQM simulations.

3. Perform critical review of Shenandoah National Park Study (summer 1991), and other intensive field studies, as data source(s) for assessing relationships between b_{sp} , RH, sulfate mass concentration and composition, and diurnal sulfate variations. Use data, as appropriate, for assessment analysis. Compare critical data needs for validating aerosol light extinction relationships with available data sources and prepare field study plan to fill in data gaps for future assessments.
4. Use NPS IMPROVE Network data base, supplemented with data from Trijonis et al. (1990) for seasonally averaged aerosol mass concentrations, to generate hourly values of chemically-specified aerosol mass concentrations (along with variance statistics) for each day of the year and use NOAA Local Climatology reports for RH and T data. Use Placet et al. (1990) for emissions estimates. Explore the use of sinusoidal diurnal aerosol and RH variations, as in the Scoping Study, with coefficients developed from intensive study periods, RADM simulations and climatological data. Estimate hourly patterns for nitrate aerosol using RADM output for gas-phase species and sulfate as input to a "box" model employing MARS.

5. Use same characterization sites as in the Scoping Report:

- Shenandoah National Park
- Grand Canyon National Park

Estimate uncertainties associated with interpolations or extrapolations to areas for which no field data are available.

6. Classify computed aerosol light extinction coefficients and visual ranges for each case (base and 1.2, 0.6, and 0.2 X base) in terms of frequency and/or probability distributions for site during daylight hours and present results graphically (bar charts, probability graphs) along with photographs for each site and each class. Adjust results for inclement weather conditions (precipitation, fog, blowing dust, etc.).

Future Assessments

Future assessments will develop more realistic simulations of diurnal and daily aerosol mass concentration behavior by augmenting available monitoring data with model simulations. Data from selected intensive periods will continue to be used to validate the daily and diurnal behavior derived from the model simulations. Improvements in the relationships between aerosol light-scattering coefficient and aerosol mass concentration and relative humidity, as developed from intensive field studies and Mie code sensitivity studies, will also be used to improve the assessment results. Also, realistic representations of non-energy background visibility impairment will be included with the emission change scenarios to assess the realism of detecting visibility improvement with respect to emission changes. Finally, the assessments will examine the geographic variability of visibility impairment and emission changes by considering additional characterization sites than were included in the Scoping Study and the 1992 Assessment. The elements of the future assessments are expected to be as follows:

1. Tools/Relationships

a) M_{aer} vs emissions:	Assume linear relationship, but use VAQM simulations to enhance predictive capability of assessment method.
b) b_{ext} vs M_{aer} :	Refined relationships based on additional field studies, especially size-resolved chemical composition and application of Mie theory.
c) b_{ext} vs RH and composition:	MARS or SEQUILIB models in VAQM.
d) VAQ Index vs b_{ext} :	Use L_v .

2. Test refined SEQUILIB, MARS, and/or other models with state-of-the-art descriptions of secondary aerosol formation for inorganic and organic aerosols with high temporal resolution temporal data from additional new field studies to be conducted (see below).

Calculate aerosol light-scattering coefficient (b_{sp}) from particle composition and water content using refined relationships derived in related DOE or other agency projects and compare to available nephelometer-measured values of b_{sp} obtained at dry and ambient RH conditions. Continue testing of VAQM and associated aerosol optics models with new data or data from other studies.

Use refined aerosol models and refined extinction algorithms to estimate hourly b_{sp} values for an entire annual cycle from available seasonally averaged aerosol mass concentration data with adjustments for RH and T based on seasonal and diurnal climatological information (see below). Estimate diurnal variations in aerosol mass concentrations from recent field study or other data sources and factor these variations into the calculations. Repeat the calculations using dry aerosol mass concentrations that are scaled by factors of 1.2, 0.6, and 0.2. Repeat calculations using VAQM simulations to estimate sulfate and other secondary aerosol response to emission changes and to enhance predictive capability of assessment method.

3. Prepare newly acquired data and continue to review data from all other recent field studies, such as Shenandoah National Park (NPS, summer 1991), Uniontown (EPRI, summer 1990) and Hopi Point (SRP, winter 1990), for use in evaluating relationships between b_{sp} , RH, sulfate mass concentration and composition, and diurnal sulfate variations. Use data, as appropriate, for evaluations. Continue to compare critical data needs for validating aerosol light extinction relationships with available data sources and prepare an augmented field study plan to fill remaining data gaps for future assessments.
4. Use NPS IMPROVE Network data base, supplemented with data from Trijonis et al. (1990) for seasonally averaged aerosol mass concentrations, to generate hourly values of chemically speciated aerosol mass concentrations (along with variance statistics) for each day of the year and use NOAA Local Climatology reports for RH and T data. Use Placet et al. (1990) for emissions estimates. Explore the use of sinusoidal diurnal aerosol and RH variations, as in the Scoping Study, with coefficients developed from intensive study periods, new VADM simulations and climatological data. Estimate hourly patterns for nitrate and organic aerosol using VADM output for gas-phase species and sulfate as input to "box" models of inorganic (MARS or SEQUILIB) and organic aerosols.
5. Expand the number of characterization sites from those used in the Scoping Report and 1992 Assessment for better representation of national visibility impairment conditions.
6. Classify computed aerosol light extinction coefficients and visual range each case (base and 1.2, 0.6, and 0.2 X base) according to frequency of occurrence at each site during daylight hours and present results graphically (bar charts, probability graphs) along with photographs for each site and each class. Adjust results for inclement weather conditions (precipitation, fog, blowing dust, etc.). Provide estimate of uncertainty in results arising from measurement variability and parameter uncertainty.

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Appendix

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September 19-20, 1991

Boulder, Colorado

Appendix

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September 19-20, 1991

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