CHAPTER 1

INTRODUCTION

In Section 169A of the Clean Air Act as amended August 1977, Congress declared, as a national goal, "the prevention of any future, and the remedying of any existing, impairment of visibility in mandatory Class I Federal areas which impairment results from manmade air pollution." Mandatory Class I Federal areas are national parks larger than 6000 acres, wilderness areas larger than 5000 acres, and international parks regardless of size, all of which were in existence on August 7, 1977. There are 158 Class I areas, of these areas 156 have been identified as having visibility related attributes that require protection.

This section of the Clean Air Act required the Environmental Protection Agency (EPA) to promulgate regulations requiring states to incorporate Class I area visibility protection in their State Implementation Plans (SIPs). These EPA regulations, promulgated on December 2, 1980, included a section requiring the states to develop a monitoring strategy for evaluating visibility in the mandatory Class I areas and to use monitoring data in decisions required by the visibility protection program. On July 12, 1985, EPA promulgated federal regulations for states that did not submit visibility SIPs.

The 1980 EPA regulations called for the establishment of a cooperative visibility monitoring effort between the EPA and several federal land management agencies: the National Park Service (NPS), the Fish and Wildlife Service (FWS), the Bureau of Land Management (BLM), and the Forest Service (FS). In 1991 several additional organizations joined the effort: State and Territorial Air Pollution Program Association (STAPPA), Western States Air Resources Council (WESTAR), and Northeast States for Coordinated Air Use Management (NESCAUM).

This cooperative visibility monitoring effort was named IMPROVE, for the Interagency Monitoring of Protected Visual Environments.

This report is the first in a series of annual reports that describe the data collected by the IMPROVE monitoring network. The objectives of this report are three-fold:

(1) To describe the spatial and temporal variation of visibility, as measured by the light extinction coefficient, and the chemical composition of the visibility-degrading aerosol[1] for the first three years of operation of the network: Spring 1988 through

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[1] An aerosol is a suspension of fine and coarse solid and liquid particles in air. Particles, especially fine particles less than 2.5 μm, scatter light and degrade the visual information content of a scene (e.g., contrast, color, line, and texture).

Winter 1991;

(2) To provide a first estimate of the apportionment of visibility impairment to the fundamental chemical species, such as sulfates, nitrate, organics and elemental carbon, and soil dust; and

(3) To compare measurements of light extinction to calculations of light extinction reconstructed from the component chemical species.

1.1 Objectives of Visibility Monitoring

The primary objectives of IMPROVE are the following:

(1) To establish current background visibility levels in Class I areas;

(2) To identify chemical species and emission sources responsible for existing man-made visibility impairment; and

(3) To document long-term trends for assessing progress toward the national visibility goal.

By measuring visibility routinely over a network and over a sufficiently long period of time, the first and third objectives of IMPROVE can be met. The monitoring also meets a portion of the second objective: the identification of the chemical composition of the visibility-degrading aerosol.

Each of these IMPROVE objectives are discussed in greater detail below.

Establish Current Visibility. This is necessary for two reasons. First, visibility levels monitored at a Class I area, when compared to surrounding area visibility or area estimates for natural levels, may be sufficient to indicate man-made impairment. Second, knowledge of existing visibility levels is required to model the anticipated visibility effects of proposed emission sources, because increments of pollution are more noticeable in clear conditions.

Establishment of present visibility levels requires monitoring which is appropriate for both surface and elevated layer impairment distributions. Optical monitoring systems, such as the transmissometer, are appropriate for surface haze monitoring, while scene monitoring with photography is the only practical way to routinely monitor elevated layers.

Fine particles consist of different chemical species either within the same particle (internally mixed) or in different particles (externally mixed). Significant chemical species found in particles include sulfates, nitrate, organic and elemental carbon, and soil dust. The sulfates, nitrate, and some hygroscopic organics absorb water from the atmosphere, thereby increasing significantly the light-scattering particle size and mass.

Visibility changes with time: diurnal, seasonal, and yearly variations all exist. Though five to eight years of data would be considered ideal for establishing present seasonal and annual averaged conditions, a minimum of one year is a reasonable compromise if that year is typical from a meteorological and source activity point of view.

Source identification. Identification of chemical species and emission sources responsible for man-made visibility impairment is necessary to protect Class I areas, as called for by Congress. Monitoring is the principal means of gathering information needed to identify the contribution to impairment by emission sources. Even to distinguish man-made from natural impairment, which is fundamental to the national visibility goals, requires information derived from monitoring data.

Aerosol and scene monitoring are the primary sources of emission source identification information. Photography of a plume emanating from its source and impacting a Class I area is sufficient to indicate impairment. Further, photographs can be evaluated to indicate the density or intensity of the visible plume. Unfortunately, most visibility impairment does not lend itself to this simple type of source attribution. Often sources are not visible from any line of sight that includes the Class I area, or their plumes disperse to a haze layer before reaching it.

Visibility impacts are often caused by aerosols formed over time from gaseous pollutants that are emitted without visibly noticeable plumes. Characteristics of the aerosol that are responsible for the haze provide valuable information that can be used in conjunction with other information to help identify the responsible emission sources. It is possible to statistically relate measured optical data to corresponding aerosol composition data to estimate the relative importance of the various major components of the aerosol. The result, known as an extinction budget, should narrow the list of possible sources responsible for large impacts. For example if organic carbon is shown to be responsible for 75% of the extinction coefficient, the major sources responsible must emit organic carbon.

Another related approach for source identification using aerosol data is known as receptor modeling. Instead of using only the major aerosol components that are directly responsible for the impairment, receptor models use relative concentrations of trace components which can more specifically identify the influence of individual sources (or source types).

Long-term trends. With the establishment of a long-term goal of no man-made visibility impairment in protected areas, Congress imposed the responsibility to show progress towards meeting that goal. Trends monitoring is an ideal approach for tracking the visibility conditions of Class I areas.

Optical and scene monitoring conducted to establish present visibility levels (described above), if conducted in perpetuity, will provide the data required to determine long-term visibility trends. In order to determine the effectiveness of individual concurrent emission reduction programs, it is necessary to conduct periodic aerosol monitoring to support extinction budget analysis as described above.

1.2 Overview of the IMPROVE Monitoring Network

The design of the IMPROVE monitoring network was resource and funding limited so that it was not practical to place monitoring stations at all 156 mandatory Class I areas where visibility is an important attribute. Instead, the IMPROVE Steering Committee selected a set of sites that were representative of the Class I areas. A total of 36 sites (20 IMPROVE and 16 NPS/IMPROVE protocol sites) are examined in this report. Each has aerosol monitoring and scene monitoring equipment (automated cameras); however, only 20 of the sites have optical monitoring equipment (e.g., transmissometers or nephelometers to measure visibility related parameters).

Figure 1.1 shows a map of the United States showing the locations of the 36 monitoring sites analyzed in this report. On the basis of regional similarities, the sites were grouped into 19 regions, listed in Table 1.1.

The routine IMPROVE monitoring approach involves aerosol, optical, and view monitoring. Aerosol monitoring measures the mass concentration (in micrograms per cubic meter, $\mu g/m^3$) and the chemical composition of the particles. Optical monitoring measures the light extinction coefficient (b_{ext}) using a transmissometer or the light scattering coefficient (b_{scat}) using a nephelometer. View monitoring documents the appearance of the scene by automated photography using color slide film.

Aerosol monitoring in the IMPROVE network is accomplished by a combination of particle sampling and sample analysis. The sampler employed was designed specifically for the program. It collects four simultaneous samples: one PM_{10} sample (particles less than 10 micrometers, μm, in diameter) on a Teflon filter and three $PM_{2.5}$ samples (particles less than 2.5 μm in diameter) on Teflon, nylon, and quartz filters. Each of the four samples is collected by a separate subsystem (or module) including everything from the inlet to the pump with only the support structure and controller/timer in common. The particle size segregation for the PM_{10} module is accomplished by a wind insensitive inlet with a 10 μm cutoff, while the $PM_{2.5}$ segregation is produced by passing the sampled air through a cyclone separator. Constant sample flow is maintained by a critical orifice in each module. The IMPROVE sampler is programmed to automatically collect two 24-hour duration samples per week.

Only mass analyses are conducted on the PM_{10} samples. The $PM_{2.5}$ samples are analyzed for mass, elements, ions (including particulate nitrate sampled through a denuder), organic and elemental carbon, and optical absorption.

At most sites in the IMPROVE network, long-path transmissometers are employed for optical measurements. These instruments measure the amount of light transmitted through the atmosphere over a known distance, usually 0.5 to 10 kilometers, between the light source (transmitter) and the light monitoring component (receiver). Transmission measurements are converted electronically to the path-averaged light extinction coefficient (b_{ext}). At a few sites nephelometers are used which internally measure the light scattering coefficient (b_{scat}).

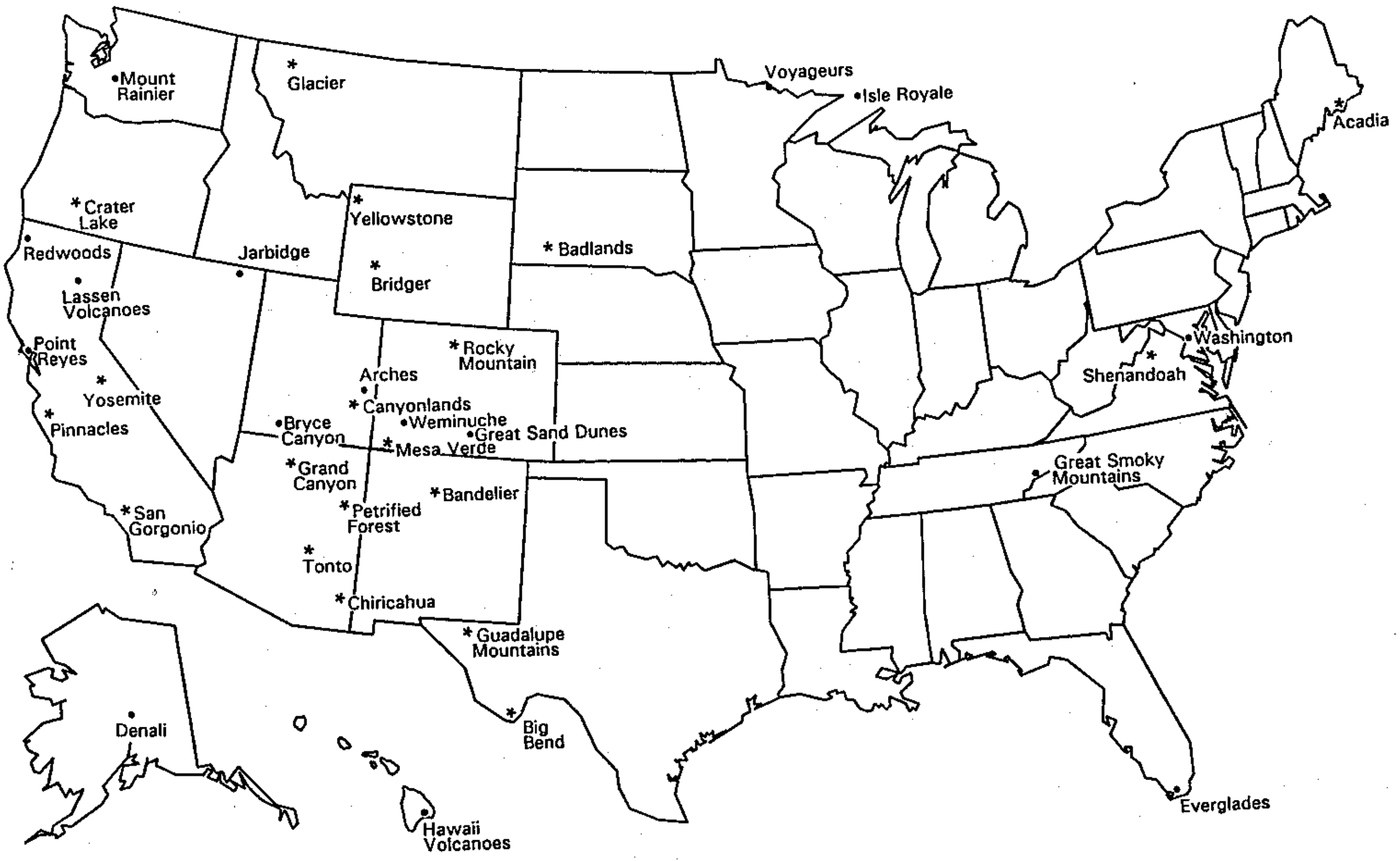

Figure 1.1. The 36 IMPROVE sites included in the report. Sites designated by an asterisk have both aerosol and visibility monitoring.

Table 1.1 IMPROVE and NPS/IMPROVE protocol sites according to region.

Alaska (AKA)
- Denali National Park (DENA)

Appalachian Mountains (APP)
- Great Smoky Mountains National Park (GRSM)
- Shenandoah National Park (SHEN)

Boundary-Waters (BWA)
- Isle Royale National Park (ISRO)
- Voyageurs National Park (VOYA)

Cascade Mountains (CAS)
- Mount Rainier National Park (MORA)

Central Rocky Mountains (CRK)
- Bridger Wilderness Area (BRID)
- Great Sand Dunes National Monument (GRSA)
- Rocky Mountain National Park (ROMO)
- Weminuche Wilderness Area (WEMI)
- Yellowstone National Park (YELL)

Coastal Mountains (CST)
- Pinnacles National Monument (PINN)
- Point Reyes National Seashore (PORE)
- Redwood National Park (REDW)

Colorado Plateau (CPL)
- Arches National Park (ARCH)
- Bandelier National Monument (BAND)
- Bryce Canyon National Park (BRCA)
- Canyonlands National Park (CANY)
- Grand Canyon National Park (GRCA)
- Mesa Verde National Park (MEVE)
- Petrified Forest National Park (PEFO)

Florida (FLA)
- Everglades (EVER)

Great Basin (GBA)
- Jarbidge Wilderness Area (JARB)

Hawaii (HAW)
- Hawaii Volcanoes National Park (HAVO)

Northeast (NEA)
- Acadia National Park (ACAD)

Northern Great Plains (NGP)
- Badlands National Monument (BADL)

Northern Rocky Mountains (NRK)
- Glacier National Park (GLAC)

Sierra Nevada (SRA)
- Yosemite National Park (YOSE)

Sierra-Humboldt (SRH)
- Crater Lake National Park (CRLA)
- Lassen Volcanoes National Park (LAVO)

Sonoran Desert (SON)
- Chiricahua National Monument (CHIR)
- Tonto National Monument (TONT)

Southern California (SCA)
- San Gorgonio Wilderness Area (SAGO)

Washington, D.C. (WDC)
- Washington, D.C. (WASH)

West Texas (WTX)
- Big Bend National Park (BIBE)
- Guadalupe Mountains National Monument (GUMO)

View monitoring is accomplished by automated 35-mm camera systems. These systems take three color slides per day to document the appearance of a selected scene at each of the IMPROVE sites. The slides are used to interpret measurements, to communicate perceived visual conditions, and, if needed, to derive quantitative estimates of light extinction by microdensitometry.

In addition to the aerosol, optical, and view monitoring, most sites have temperature and relative humidity instruments. Liquid water is a component of the hygroscopic sulfate, nitrate, and possibly organic carbon fractions, but it is not efficiently captured by filter sampling techniques. Relative humidity measurements are used to estimate the amount of liquid water associated with these particles.

1.3 Background Regarding Visibility Impairment and Aerosols

Visibility is usually characterized either by visual range (the greatest distance that a large dark object can be seen), or by the light extinction coefficient (the attenuation of light per unit distance due to scattering and absorption by gases and particles in the atmosphere). Under certain assumed conditions these two measures of visibility can be shown to be inversely related to each other. Visual range functions well as an aid in military operations and transportation safety. Issues of concern for such use include: the minimum distance required to land an aircraft, the distance to the first appearance of a military target or an enemy aircraft or ship, and safe maneuvering distances under impaired visibility conditions. Because of the use of familiar distance units, the simple definition, and the ability of any sighted person to characterize visual conditions with this parameter without instruments, visual range is likely to remain the most popular measure of atmospheric visibility.

Extinction coefficient is used most by scientists concerned with the causes of reduced visibility. There are direct relationships between the concentrations of the atmospheric constituents and their contribution to the extinction coefficient. Apportioning the extinction coefficient to atmospheric constituents provides a method to estimate the change in visibility caused by a change in constituent concentrations. This methodology, known as extinction budget analysis, is important for assessing the visibility consequences of proposed pollutant emission sources, or for determining the extent of pollution control required to meet a desired visibility condition. Interest in the causes of visibility impairment is expected to continue and the extinction coefficient will remain important in visibility research and assessment.

Neither visual range nor extinction coefficient is linear with visual scene changes caused by uniform haze (i.e., as opposed to elevated haze layers and plumes). For example, a given change in visual range or extinction coefficient can result in a scene change which is either unnoticeably small or very apparent depending on the baseline visibility conditions. Presentation of visibility measurement data or model results in terms of visual range or extinction coefficient can lead to misinterpretation by those who are not aware of the nonlinear relationship.

To rigorously determine the perceived visual effect of a change in extinction coefficient requires the use of radiative transfer modeling to determine the changes in light from the field of view arriving at the observer location, followed by the use of psychophysical modeling to determine the response to the light by the eye-brain system. Results are dependent not only on the baseline and changes to atmospheric optical conditions, but also on the characteristics of the scene and its lighting. The complexity of employing such a procedure and the dependence of the results on non-atmospheric factors prevent its widespread use to characterize perceived visibility changes resulting from changes in air quality.

Parametric analysis methods have been used to suggest that a constant fractional change in extinction coefficient or visual range produces a similar perceptual change for a scene regardless of baseline conditions. Simplifying assumptions eliminate the need to consider the visibility effects of scene and lighting conditions. Using the relationship of a constant fractional change in extinction coefficient to perceived visual change, a new visibility index called deciview (dv) is defined as:

$$dv = 10 \ln(b_{ext} / 0.01\, km^{-1}) , \tag{1.1}$$

where extinction coefficient is expressed in km^{-1} (Pitchford and Malm, 1993). A one dv change is about a 10% change in extinction coefficient, which is a small but perceptible scenic change under many circumstances. The deciview scale is near zero for pristine atmosphere (dv = 0 for Rayleigh condition at about 1.8 km elevation) and increases as visibility is degraded. Like the decibel scale for sound, equal changes in deciview are equally perceptible.

1.3.1 Relationship Between Visibility and Aerosol Concentrations

Visibility is degraded by light scattered into and out of the line of sight and by light absorbed along the line of sight. Light extinction (the sum of light scattering and absorption) is usually quantified using the light extinction coefficient (b_{ext}), which may be thought of as the atmospheric concentration of light extinction cross-sectional area. Light extinction has units of m^2/m^3 or m^{-1}.

The light extinction coefficient (b_{ext}) is the sum of the light scattering coefficient (b_{scat}) and the light absorption coefficient (b_{abs}). Light scattering results from the natural Rayleigh scatter (b_{Ray}) from air molecules (which causes the blue sky) and the scattering caused by suspended particles in the atmosphere (aerosols). Particle scatter (b_{sp}) can be caused by natural aerosol (e.g., wind-blown dust and fog) or by man-made aerosols (e.g., sulfates, nitrates, carbonaceous aerosol and other fine and coarse particles). Light absorption results from gases (b_{ag}) and particles (b_{ap}).

Nitrogen dioxide (NO_2) is the only major light absorbing gas in the lower atmosphere; its strong wavelength-dependent scatter causes yellow-brown discoloration if present in sufficient quantities. Soot (elemental carbon) is the dominant light absorbing particle in the atmosphere. Thus, the total light extinction is the sum of its components:

$$b_{ext} = b_{scat} + b_{abs} = b_{Ray} + b_{sp} + b_{ag} + b_{ap} \, . \tag{1.2}$$

The particle light scattering coefficient (b_{sp}), in turn, is composed of the contributions from individual species. Fine particles are much more efficient at light scattering (per unit mass) than larger particles. Thus, it makes sense to divide the contributions to b_{sp} into the contributions from various species of fine and coarse particles. In this study, we specifically evaluated the following components of fine particles (those with diameters less than 2.5 μm): sulfate (SO), nitrate (NO), organic carbon, elemental carbon (soot), and others. In addition to these chemical species, the effect of water associated with sulfate, nitrate, and some organics need to be considered in the overall assessment of light extinction. Finally, the coarse fraction of PM_{10} (those with diameters between 2.5 and 10 μm) and giant particles (those with diameters greater than 10 μm) are separately considered.

The light extinction coefficient can be written as the sum of the products of the concentrations of individual species and their respective light extinction efficiencies:

$$b_{ext} = b_{Ray} + \sum \beta_i \, C_i \, , \tag{1.3}$$

where β_i is the light extinction efficiency (m^2/g) of species i, C_i is the atmospheric concentration of species i ($\mu g/m^3$), and the summation is over all light-interacting species (i.e., sulfate, nitrate, organic carbon, elemental carbon, other fine particles, coarse particles, giant particles, and NO_2). The above units, when multiplied, yield units for b_{ext} of 10^{-6} m^{-1} or $(10^6 \ m)^{-1}$, or as we prefer to label it here, inverse megameters (Mm^{-1}).

1.3.2 Effect of Relative Humidity on Light Scattering

Sulfates, nitrates, and some organics can combine with water in the vapor phase to form solutions. Thus, at some humidity conditions, considerable water may be associated with these species. Although the overall light scattering efficiency is on the order of 3 m^2/g for these solutions, if the light scattering efficiency is stated in terms of the mass of dry sulfate (SO), the efficiency must be larger than 3 m^2/g to account for the additional mass (and volume) of the associated water. In addition, the associated cations (H^+ and NH) must also be included. As a result, light scattering efficiency per unit of dry sulfate can be much larger than 3 m^2/g. This hygroscopic effect can be described by the following equation:

$$\beta_{wet} = k\ f_{RH}\ \beta_{dry} \tag{1.4}$$

where β_{wet} is the light extinction efficiency of the wet sulfate, nitrate, and/or organic solution, k is the ratio in molecular weight of the neutralized species (e.g., ammonium sulfate or ammonium nitrate) to the anion (sulfate, nitrate), f_{RH} is a factor that accounts for the liquid water associated with the aerosol at the given relative humidity (*RH*), and β_{dry} is the light extinction efficiency of the dry particle.

1.4 Organization of the Report

This report is divided into eight chapters. Chapter 2 summarizes the methodologies, protocols, and uncertainties of aerosol and optical monitoring. The assumptions for determining the chemical composition of the particles are discussed in Chapter 3. Chapter 4 presents the results of various cross-checks and comparisons for quality assurance and validation of the parameters derived from the aerosol measurements. A discussion of the acidity of the sulfate component of the aerosols is also included. The spatial and seasonal patterns of aerosol mass and chemical composition are summarized in Chapter 5. Chapter 6 discusses the theory and results of light extinction apportionment by aerosol chemical species and the spatial and seasonal patterns of light extinction and reconstructed light extinction. Chapter 7 compares the reconstructed with measured extinction and describes some regional characteristics of the measured extinction.