Chapter 1

INTRODUCTION

THE DESIGN AND IMPLEMENTATION OF THE WINTER HAZE INTENSIVE TRACER EXPERIMENT -WHITEX

1.1 Overview

Protection of vistas for certain national parks and wilderness areas as provided by the Clean Air Act Amendments of 1977(1) has stimulated an interest in visibility research. Methods are being developed and used to characterize atmospheric transparency, to identify the relative importance of the various particulate and gaseous atmospheric materials and to determine the role of man-made emissions. Much of the research has been conducted in the desert Southwest, in particular in northern Arizona and southern Utah. The juxtaposition of energy resources (especially coal) and national parks (including Grand Canyon, Bryce Canyon and Canyonlands) in an area where small changes in aerosol concentration can significantly affect visibility justifies concern by government and private organizations for visibility impacts resulting from industrial emissions. Figure 1.1 is an emission density map showing locations of major *SOx* sources and national parks on the Colorado Plateau.

Accordingly, a cooperative effort, the Subregional Cooperative Electric Utility (comprised of the Electric Power Research Institute, Southern California Edison and the Salt River Project), National Park Service (NPS), Environmental Protection Agency (EPA) and Department of Defense (DOD) Study, SCENES, is centered in this area. It operates on a five-year plan (1984-1989) involving continual visibility and aerosol measurements at a dozen locations, plus more in-depth intensive and special studies conducted over shorter, seasonally representative periods. One of these, the Winter Haze Intensive Tracer Experiment (WHITEX) was conducted in January and February 1987 in the Colorado River area of the Colorado Plateau.

1.2 Background

The Colorado Plateau, with its many associated class I national park areas, was chosen as the location to implement a scoping study designed to evaluate the ability of a variety of receptor modeling approaches to attribute visibility impairment in a number of class I areas to emissions from a single point source, the Navajo Generating Station. The area, shown in Figure 1.3, is by most standards remote, undeveloped and sparsely populated. The nearest large urban areas are over 300 km away. Only a few smaller urban areas or towns are within the area, these include Moab, Utah, Page, Arizona and at the most western end of the study area, Las Vegas, Nevada.

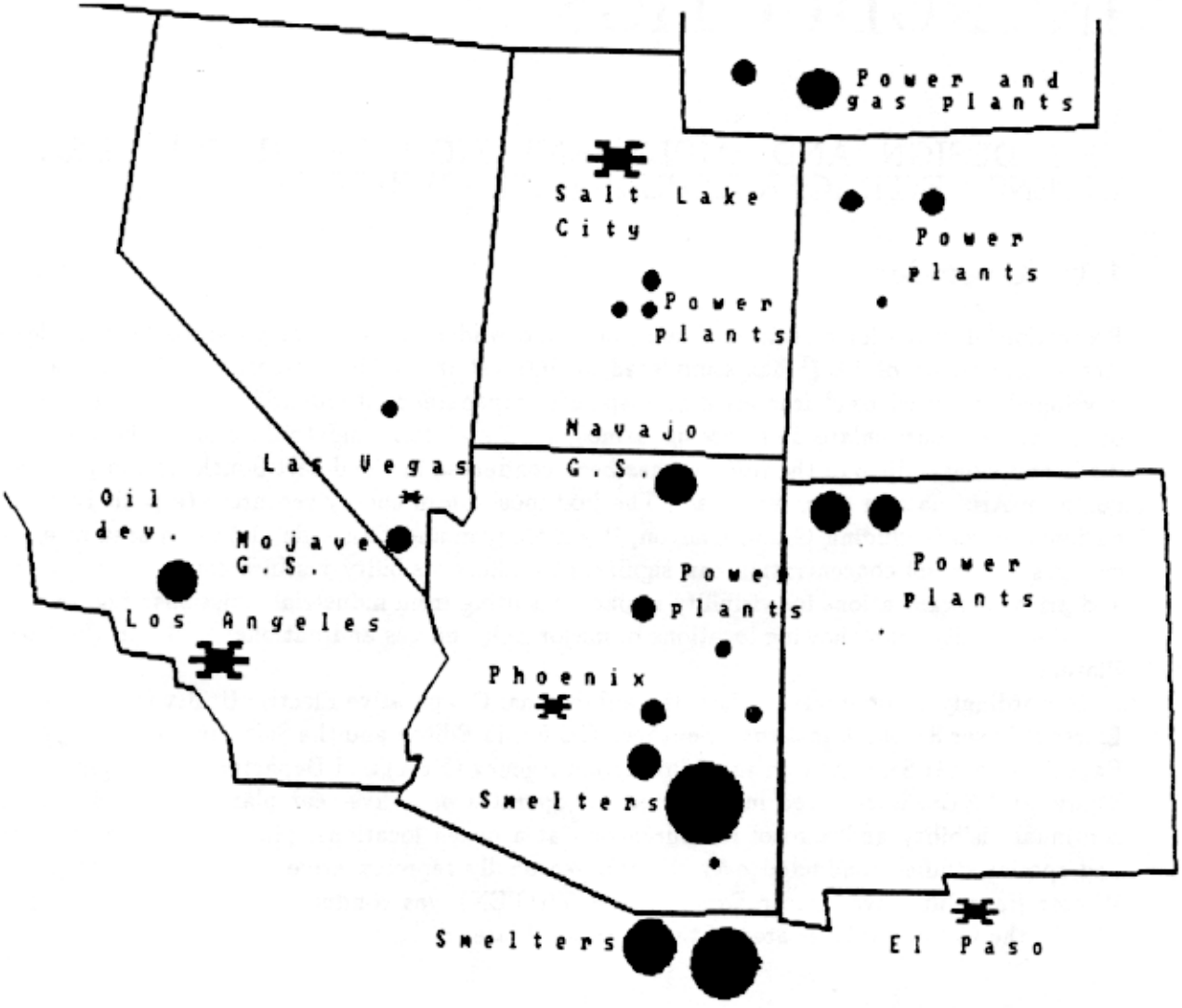

Figure 1.1: Approximate SO_x emissions from major point sources and urban areas in the southwest United States for 1987.

There are a few small industrial enterprises in the vicinity such as sawmills, mining and milling operations and a number of large coal fired electric generating facilities, one of which is the Navajo Generating Station (NGS) located near Page, Arizona. The Navajo Generation Station is a large (2300 MWe) coal-fired power plant without sulfur dioxide control equipment; thus, it is a significant single contributor of sulfur species -sulfur dioxide *(S02)* and sulfate particles *(S04)* -to the atmosphere of the region. With the control and shutdown of several smelters in the western U .S., NGS has become the largest single S*02* emission source in the West.(2)

The terrain surrounding the lower Colorado River rises to about 800 meters above the water's surface. Wintertime meteorology in the area is characterized by several periods of stagnation, of about one week each. Air pollutants can be trapped by a persistent thermal inversion below the height of the surrounding terrain during the stagnation periods, resulting in a distinct visible surface haze layer. These stagnant periods are interrupted by synoptic-scale fronts with associated strong winds sweeping rapidly through the area.(3) The winter haze over the Colorado Plateau area has been routinely documented with photographs since 1978 by the EPA, NPS and BLM (Bureau of Land Management). The haze is usually seen as a bright white layer with a distinct upper edge and it occasionally includes one or more perceptible layers.(3, 4)

A number of earlier investigations have been prompted by the need to determine the origins of the haze. NPS sponsored several modeling efforts to evaluate the possibility that the Navajo Generating Station is partially responsible for the haze.(5, 6) A wind field model was adapted for the area's terrain and winter meteorology to investigate transport and diffusion. In a separate study, ambient nitrogen chemistry was theoretically simulated to estimate the role of particulate nitrates.(7) Though both efforts added to the knowledge of the source of the haze, the uncertainties in modeling this situation led to approaching the question with observational studies.

A SCENES special study was previously conducted in 1986 at Glen Canyon to provide informa- tion to aid in planning subsequent observations.(8) The primary objectives of this exploratory study were to determine the horizontal and vertical extent of the haze and to identify major constituents of the haze. Aircraft-based measurements confirmed the haze to be more extensive horizontally than just the Glen Canyon area (e.g., extending at least to Bryce and Grand Canyon to the west). This greater horizontal extent enlarges the number of possible emission sources to be considered. This complicates the source attribution because the contribution of any one source may vary considerably with time and location within the haze. In mapping the vertical extent of the haze with an instrumented aircraft, the air above the inversion layer was found to be essentially particle-free, while below the layer, scattering coefficients varied from two to five times clean air values. Sampling at the south end of the lake only, the particles were found to be composed largely of sulfates and organics. Nitrates were found to be primarily gaseous, with about a tenth of the total being particulate nitrate.

1.3 Study Plan

Shortly after the completion of the winter 1986 study, the SCENES participants began planning a more comprehensive effort for the winter of 1987 to address persistent questions about the nature and sources of winter haze conditions. The overall study objective was to assess the feasibility of attributing emissions from a single point source to visibility impairment in prespecified geographic regions. Specifically, various receptor and deterministic models were to be evaluated and intercompared as to their ability to link Navajo Generating Station emissions to visibility impairment at Grand Canyon and Canyonlands National Park and Glen Canyon National Recreation Area. Meeting this objective is a three tier process. First the relative contribution at the receptor site

of primary and secondary aerosols associated with the NGS must to be achieved. Secondly, the contribution of these aerosol species to atmospheric optical variables needs to be established, and finally the contribution of optical variables associated with power plant emissions to an incremental decrease in visibility below that which would have existed otherwise needs to be examined.

The major focus of the WHITEX study is centered around the evaluation of receptor oriented approaches for linking NGS emissions to aerosol concentrations in Grand Canyon and Canyonlands National Parks and Glen Canyon National Recreation Area. In a receptor type model the atmosphere is treated as a black box through which source emissions are transferred to the receptor site. Source receptor relationships are empirically developed using statistical inference techniques. Historically, the chemical mass balance (CMB) formalism has been most often used to link source emissions to aerosol concentrations at a receptor site. The CMB approach essentially uses ratios of trace material associated with different sources in combination with trace material measured at the receptor site to apportion primary (nonconverting) aerosol species. However, CMB has serious short comings in that it is not designed to apportion secondary aerosols, such as ammonium sulfate, to its *S02* source. Other common types of receptor models include principal component analysis (PCA) and multiple linear regression (MLR). Explanations of these models are given by Watson,[9] Chow,[10] and Hopke.[11] Furthermore, Dzubay et al.,[12] Lewis and Stevens,[13] and Stevens and Lewis[14] have integrated a number of these approaches into a hybrid receptor model that can be used to apportion secondary aerosols. All these models can be shown to be special cases of a deterministic statement, referred to in this report as the general mass balance model (GMB), of how gases and aerosols are transported and transformed as they pass through the atmosphere. The GMB model is discussed in detail in Appendix 6A.

In this report a regressional model, derivable from the GMB equations and referred to as the tracer mass balance regression (TMBR) model, will be used to apportion secondary aerosol species. A full derivation of TMBR can be found in Appendix 6B. TMBR is formulated to apportion secondary aerosols if certain assumptions are met. First, a unique trace material must be associated with a source or group of sources and secondly the atmospheric transfer processes must be approximated be a linear model. If a unique tracer is not available CMB can be used to apportion non-unique tracer species to source types and the analysis can still be carried out. TMBR essentially relies on relative changes in secondary aerosol and tracer concentrations over time to yield the desired apportionment.

Finally, a differential mass balance model (D MB) having elements of both deterministic and receptor modeling approaches will be used to apportion secondary aerosols. The term differential is derived from the use of unique trace material spatial concentration gradients to calculate atmospheric dispersion. Atmospheric deposition, chemical conversion and transport time are calculated from first principles. A full derivation of the D MB model can be found in Appendix 6C.

Table 1.1 outlines the different approaches as well as summarizes the major advantages and disadvantages of each technique. For the sake of completeness, advantages and disadvantages of deterministic modeling are also included in Table 1.1.

Several less quantitative approaches are also used to gain insight into basic physio-chemical processes at work over the time period for which apportionment estimations are carried out. These include evaluation of the relative emission strengths, plume trajectory and streakline analysis, spatial and temporal trends, analysis of synoptic meteorological conditions, and deterministic wind field modeling on the mesoscale level (<200 km).

The focus of attributing NGS emissions to optical variables will be directed toward the relationship between various attributed aerosol species and optical extinction. Since a change in atmospheric transmittance (extinction) under a variety of conditions has been shown to be a good approximation to the change in the atmospheric modulation transfer function.(15) The optical ex-

Table 1.1: Brief description of the advantages and disadvantages of various receptor techniques examined in the Winter 1986 WHITEX study.

RECEPTOR MODELING METHOD	DESCRIPTION	ADVANTAGES	DISADVANTAGES
Tracer Mass Balance Regression (TMBR)	Passive tracer release from point source over extended period of time. Measurement of tracer and aerosol species at receptor site over time allows for building a regression model relating aerosol species to point source emissions.	Allows for establishing fractional contribution of point source emissions to various aerosol species relative to other sources and background.	Unique tracer for each source of interest may not be available. Collinearity between tracer material may lead over or underestimation of source contribution
Chemical Mass Balance (CMB)	Measurement or knowledge of source natural tracer profile along with measurement of some natural tracers at receptor sites.	Yields fractional contribution of each source to various aerosol species at receptor site.	Model requires conservation of mass over transport distance of interest. Requires all source profiles to be known. Natural tracer profiles from different sources may be similar .
Differential Mass Balance (DMB)	Passive tracer release from point source with measurements of aerosol and tracer concentrations at receptor site.	Yields insight into chemical conversion rates and deposition velocities. Yields fractional contribution of point source emissions to aerosol species at receptor site.	Results are confounded by uncertain knowledge of background aerosol concentrations and relative importance of deposition and conversion rates.
Deterministic Model Calculations	Deterministic model calculations of temporal and spatial aerosol concentration gradients are compared directly to field measurements of those same aerosol species.	Historically, experience offers a basic understanding of the chemical and physical processes involved in the production and dispersion of various aerosol species.	Requires validated models of wind fields and chemistry modules. No widely accepted models at this time.

tinction associated with particles can be calculated using Mie theory if the particle characteristics are well documented. The size distribution, shape, density and refractive index of the particles are needed for such a calculation. This information is generally either unavailable or available with insufficient detail, so that Mie theory must rely upon a number of assumptions. Mass size distribution data can be obtained using size segregating samplers, however, the capability of such samplers to correctly represent the particulate nitrate or the labile fraction of organic carbon is questionable. For instance, the mass of the most common labile component, water, is not accounted for.

A second approach for attributing aerosol species to extinction is a statistical methodology that relies on multilinear regression (MLR) analysis where it is assumed that the relationships between atmospheric extinction, *bext,* and aerosol species mass concentrations, *mi,* are represented by $bext = \sum_i \sigma_i m_i$. The measured $m_i's$ are assumed to be independent variables, *bext* is the dependent variable and regression coefficients, $\sigma_i's$, are interpreted as extinction or scattering efficiencies depending on whether *bext* or b_s is used in the analysis.

For the relationship between *bext* and the $m_i's$ to be linear requires many restrictive assumptions. Assumptions associated with inherent unknown and physical processes are discussed in some detail by White.[19] To minimize uncertainty in estimated extinction efficiency, MLR is compared to efficiencies derived from other studies.

Two ways of apportionment of extinction will be examined in this report, first by fine mass:

$$\boldsymbol{bext} = a_1 m_f + \boldsymbol{a_2\, Ce} + \boldsymbol{a_3\, [NO_2]} + \boldsymbol{b_{RAY}} \tag{1.1}$$

where m_f is fine mass associated with scattering, C_e is elemental carbon which is primarily a particle absorption term, $[NO_2]$ is nitrogen dioxide concentration, and b_{RAY} is scattering due to atmospheric gases. The second procedure is apportionment by aerosol/ chemical species:

$$\boldsymbol{bext} = \boldsymbol{a_0} + \boldsymbol{a_1\, S} + \boldsymbol{a_2 N} + \boldsymbol{a_3\, C_0} + \boldsymbol{a_4\, C_e} + \boldsymbol{a_5\, [NO_2]} + \boldsymbol{a_6\, Soil} + \boldsymbol{a_7\, R} \tag{1.2}$$

where *S, N,* C_o, C_e are particulate ammonium sulfate, ammonium nitrate, organic and elemental carbon respectively, soil is the fine mass oxides associated with *Ca, Si, Fe, K* and *Ti,* and *R* refers to mass between 2.5 μ and 10 μ .

Once source emissions have been attributed to extinction it is possible to estimate whether those emissions will effectively reduce the ability to see a landscape feature. Two optical variables which effectively characterize the visual effect of atmospheric haze on vistas are the change in contrast of adjacent scenic features or those features against the horizon sky as a function of aerosol concentration and/or composition and the contrast of the haze itself as seen against the sky or terrain background. Calculation of the change in contrast of adjacent scenic features as a function of aerosol concentration in the most general case requires a knowledge of the atmospheric modulation transfer function, $M_{tf,a}$ which, in turn, requires information on inherent scene brightness, path radiance and atmospheric transmittance between scene and observer. However, as stated above, atmospheric modulation transfer can be approximated by atmospheric transmittance under a wide variety of conditions.[21] Under these circumstances,

$$\frac{dCr}{Cr} = -Rdbext \tag{1.3}$$

where *dCr/Cr* is the percentage change in apparent contrast of a vista at a distance *R* and db_{ext} is the incremental change in extinction coefficient derived from the extinction attribution portion of the program. Thus, Equation 1.3 can be used to assess the amount of vista contrast reduction associated with NGS emissions. The calculation is straightforward and can be carried out by the interested reader. However, these calculations will not made as part of this report.

1.4 Interrelationships Between Aerosol, Optical and Visibility Apportionment

The interrelationship between measurements and apportionment methodologies as designed for the WHITEX program are schematically represented in Figure 1.2. For purposes of understanding the relative accuracy of the various receptor modeling approaches, the study was designed to calculate aerosol and optical apportionment in at least two separate ways. In this way, a relative accuracy of each technique can be estimated. For instance, measurement of *bext* and aerosol composition are combined in an MLR model to apportion extinction to aerosol species. An independent analysis using a literature review of theoretically derived extinction efficiencies allows for an independent estimation of extinction apportionment. The two estimations can then be intercompared, and differences, if any, reconciled. Similarly, tracer (CD_4) released over time can be used in a TMBR analysis to apportion sulfur and nitrate aerosols to NGS emissions, while DMB analysis will yield an independent estimation of NGS emission contribution to secondary aerosols at the receptor site. The NGS sulfate and nitrate contribution to total particulate matter at the receptor sites is then combined with the extinction apportionment analysis to yield the extinction that can be attributed to NGS. The two techniques can then be intercompared and differences, if any, reconciled. Finally, the extinction apportionment data can be combined with information from radiative transfer calculations and radiance measurement program to apportion visibility impairment.

1.5 Measurement Program

The measurement program consisted of four different types of ground station configurations and one airborne platform. The configurations are classified as major receptor, satellite, gradient, and background sites. Table 1.3 summarizes the variable measured, the methodology used to collect the data and the frequency at which the measurement was made while Table 1.2 summarizes the function of each monitoring site. Figure 1.3 shows the location of each monitoring site.

Major receptor (Type A) sites had all those measurements required for aerosol, extinction, and visibility impairment attribution while satellite sites consisted only of trace element, wind speed, and wind direction measurements. Satellite sites were used to characterize air masses flowing into and out of the study region and were used to explore temporal and spatial trends. At gradient and background sites b_{scat} fine mass, ions, carbon, trace elements, and tracer concentrations and meteorological variables were measured. Gradient sites were also used to examine spatial and temporal trends while the background site helped characterize air masses on a regional scale.

A full description of how each parameter was measured is discussed in chapter three. Therefore only a brief description of the measurements will be presented here. Atmospheric extinction was measured with a newly developed long path transmissometer.[20] Atmospheric scattering was measured with MRI 1550 integrating nephelometers which were zeroed with clean air every few hours and span calibrated twice during the course of the study.[22] Haze, contrast, and $M_{tf,a}$ can be calculated from reconstructed radiance fields derived from slides taken during the course of the study.[23] The color slides were taken using automatic photographic monitoring instrumentation comprised of 35 mm cameras using 135 mm lenses and loaded with Kodachrome 25 color slide film.

Particulate measurements were made by the IMPROVE sampler[24] at nine sites and by the stacked filter unit[25] (SFU) at three additional sites. At three of the twelve sites, the size-classifying isokinetic sequential aerosol sampler[26] (SCISAS) collected fine and total (smaller than $15\mu m$ samples on four filters. The SCISAS sampler was used primarily to establish the relative accuracy and precision of the various sampling systems.

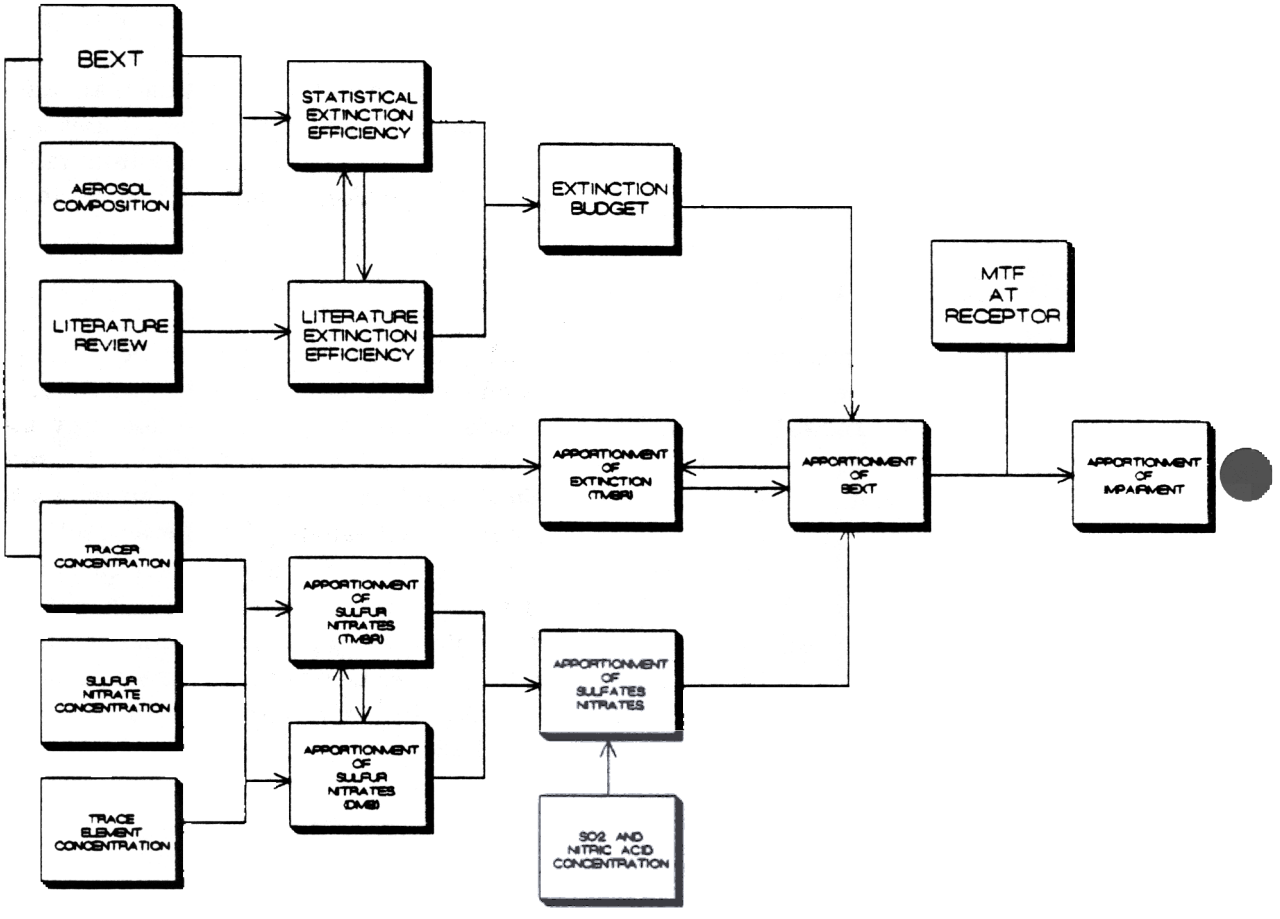

Figure 1.2: Flow diagram showing the relationship between measurement and apportionment of visibility impairment.

A "DRUM" sampler was operated to yield size resolved atmospheric element concentrations.[29] The version used for this study consisted of six rotating drums of the Mercer impactor type and to low pressure rotating drums. The nominal size ranges (aerodynamic diameter in μm) for five stages are 0.6-1.2,1.2-2.4,2.4-4.85,4.85-9.6,9.6-16.0 μm.

The optical absorption of particles on the fine Teflon filter were measured by a laser integrating plate method (IPM). Light of a 633 nm wavelength from the *He(Ne*) laser was diffused and collimated to provide a uniform beam of around 0.7 cm^2 at the sample. The light transmitted through the sample was collected with an ORIEL photodiode detection system.[30]

The tracer injected into NGS stacks was deuterated, or "heavy", methane, CD_4.[31] Though chemically similar to normal methane, heavy methane has a higher molecular weight which allows it to be distinguished by mass spectrometry at very low concentrations (1 part in 10^7). CD_4 was released continuously in proportion to NGS emissions in the time period of study. At receptor sites, 60 liters of air were pumped into large mylar/polyethylene bags and then concentrated under pressure into steel pressurized containers. The containers were shipped to Los Alamos National Laboratory for mass spectrometer analysis.

Meteorological variables were gathered using standard sensors, while upper air winds, temperature and dew point temperature were gathered using airsondes. The aircraft was equipped with an $S0_2$ monitor, an integrating nephelometer, a particulate monitor used for NGS plume characterization and bottles used for collecting large volumes of air required for the CD_4 analysis.

Table 1.2: List of the function associated with each of the monitoring sites.

SITE	SITE TYPE	FUNCTION
Glen Canyon National Recreation Area Grand Canyon National Park Canyonlands National Park	Receptor site	Calculation of the relative contribution of each aerosol species associated with NGS emissions.
Lake Mead National Recreation Area Wupatki National Monument Navajo National Monument Monticello, Utah Cisco, Utah Hanksville, Utah	Satellite	Characterize the inflow/outflow of tracers of opportunity and spatial and temporal trends
Hite, Utah Bullfrog, Utah	Gradient	Characterize spatial and temporal trends.
Bryce Canyon National Park	Background	Used to estimate background associated with long range transport from distant sources.

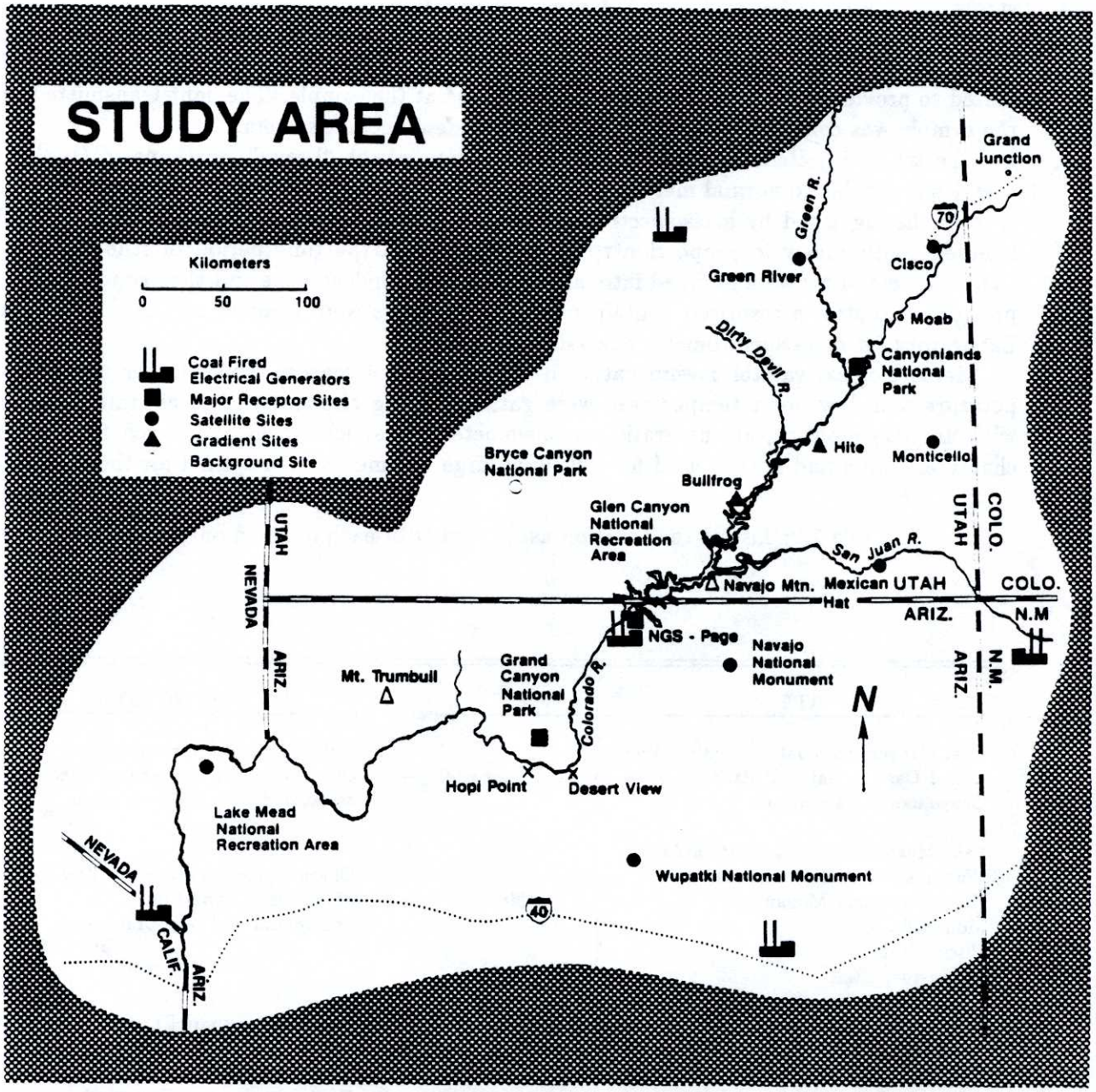

Figure 1.3: Map of SCENES WHITEX study area.

Table 1.3: List of the optical variables, aerosol species, meteorological variables, and measurement methodologies used at each of the monitoring sites.

	Location*	Method	Substrate	Frequency
Optical				
b_{SCAT}	A,C	Nephelometer	NA	Continuous
b_{ext}	A	Transmissometer	NA	Continuous
$M_{tf,a}$	A,D,E, Cider Mt., Echo Cliffs, Arches	Photographic	NA	Hourly
Haze Contrast	D, Cedar Mt., Echo Cliffs	Photographic	NA	Hourly
Particulate				
Fine Particles				
Mass	A,B,C,D	**IMPROVE/**SCISAS	Teflon	12 hr.
Ions	A,D	IMPROVE/SCISAS	Teflon	6 & 12 hr.
Nitrate (denuder)	A,D	IMPROVE/SCISAS	Nylasorb	12 hr.
Elemental & Organic Carbon	A,D	IMPROVE/SCISAS	Quartz	12 hr.
Trace Elements	A,B,C,D,E	IMPROVE/SCISAS/ **SFU	Teflon	6 & 12 & 24 hr.
Size Segregating				
Trace Elements	A	**DRUM	Teflon	6 hr.
Giant Particles (>10.0 μ)	Canyonlands	—	—	—
Large Particles (2.5–10.0 μ)	Grand Canyon, Bryce, Glen Canyon, Lake Mead	SCISAS	Teflon	12 hr.
Gases				
SO_2				
Gas chromatograph	Canyonlands	Gas Chromatograph	NA	2 & 4 hr.
K_2CO_3 Impregnated Filter	A	K_2CO_3 Impregnated Filter	Same	12 hr.
CD_4 Tracer	A	Grab Sample & Gas Chromatograph	NA	6 hr.
Meteorological				
WS,WD	A,B,C,D	—	—	10 min.
Temperature, RH	A,B,C,D	—	—	10 min.
Upper Air	Canyonlands, Glen Canyon	Airsonde	—	Twice/Day

* A = Major receptor
B = Satellite
C = Gradient
D = Background
E = Airborne Platform

** IMPROVE, SCISAS, SFU and DRUM refer to types of samplers which are discussed in some detail in the text.

1.6 Report Outline

Chapter 2 examines the climatology of the study area during the time period the WHITEX study was carried out. Chapter 3 presents the measurement program. A description of each sampling procedure and method of establishing accuracy and precision is presented. When a variable was obtained by more than one method an intercomparison of those variables is examined. Finally CD_4 injection and measurement is described. Chapter 4 is an overview of the data used when making attribution calculations. First the temporal history of each variable is presented along with pertinent descriptive statistics. Finally relationships among variables is explored with simple scatter plots of one variable plotted as a function of another. Chapter 5 examines the optical characteristics of pertinent aerosol species. Average extinction budgets are calculated for the full WHITEX time period as well as on a daily basis. Chapter 6 is the heart of the WHITEX report. The ability of various receptor modeling techniques to attribute secondary aerosols to their respective sources is explored. Chemical mass balance, tracer mass balance and differential mass balance are quantitative models investigated while several non quantitative approaches are used to either confirm or negate quantitative calculations. The non quantitative procedures were also used to gain insight into physicochemical processes at work during the study period. Appendices present a complete description of each of the receptor modeling techniques. Chapter 7 presents a deterministic calculation of wind fields and transport pathways of conservative tracers during one particular low visibility episode. The possibility of transport into and out of the study area is also explored. Chapter 8 explores the climatology of extreme (high) sulfate episodes and compares the WHITEX climatology to historic records of other sulfate episodes. Finally, Chapter 9 presents an overview of conclusions that can be drawn from the WHITEX study.

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