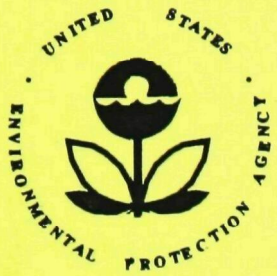

PROJECT MOHAVE

Measurement
Of
Haze
And
Visual
Effects

STUDY PLAN

September 27, 1991

UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
Office of Air Quality Planning and Standards
Research Triangle Park, North Carolina 27711

Project MOHAVE

Study Plan

U.S. Environmental Protection Agency
September 27, 1991

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List of Acronyms

ABL	Atmospheric Boundary Layer
ARS	Air Resource Specialists
BNL	Brookhaven National Laboratory
CD_4	Deuterated methane
CMB	Chemical Mass Balance
DMB	Differential Mass Balance
DRUM	Davis Rotating-drum Universal-size-cut Monitoring
ECD	Electron Capture Detector
EOF	Empirical Orthogonal Function
EPA	U.S. Environmental Protection Agency
EMSL	Environmental Monitoring Systems Laboratory
GC	Gas Chromatography
GCNP	Grand Canyon National Park
INAA	Instrumental Neutron Activation Analysis
LIPM	Laser Integrating Plate Method
LQL	Lower Quantifiable Limit
LOD	Limit of Detection
MOHAVE	Measurement of Haze and Visual Effects
MPP	Mohave Power Project
NAS	National Academy of Sciences
NGS	Navajo Generating Station
NOAA	National Oceanic and Atmospheric Administration
NPS	National Park Service
OAQPS	EPA Office of Air Quality Planning and Standards
PDCB	Perfluorodimethylcyclobutane
PDCH	Perfluorodimethylcyclohexane
PESA	Proton Elastic Scattering Analysis
PFT	Perfluorocarbon Tracer
PIXE	Particle Induced X-ray Emission
PMCP	Perfluoromethylcyclopentane
QC	Quality Control
RASS	Radio Acoustic Sounding Systems
SCE	Southern California Edison
SF_6	Sulfur hexafluoride
SO_2	Sulfur dioxide
SO_4	Sulfate
SRP	Salt River Project
TMBR	Tracer Mass Balance Regression
WHITEX	Winter Haze Intensive Tracer Experiment
XRF	X-Ray Fluorescence

1. Introduction

Reason for the Study

In 1977, in Section 169A of the Clean Air Act, Congress set 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 results from manmade air pollution." Section 169A also required EPA to promulgate regulations to assure reasonable progress toward meeting the national goal for mandatory Class I areas where visibility is an important air quality related value. On November 30, 1979, EPA identified 156 areas, including Grand Canyon National Park (GCNP), where visibility is an important air quality related value. On December 2, 1980, EPA promulgated the required visibility regulations. In broad outline, the visibility regulations require the States to coordinate their air pollution control planning activities with the appropriate Federal Land Managers to develop a program to assess and remedy visibility impairment from new and existing sources.

More recently, Congress reaffirmed its desires to address visibility issues by adding section 169B to the Clean Air Act amendments of 1990. Section 169B calls for a substantial research program to study regional haze, and requires the Administrator of EPA to establish a visibility transport commission for the region affecting the visibility of GCNP.

In January and February, 1977, the National Park Service (NPS), acting in its capacity as the Federal Land Manager for GCNP, conducted a study known as the Winter Haze Intensive Tracer Experiment (WHITEX). WHITEX involved a six-week long intensive monitoring period during which an artificial tracer was released from the Navajo Generating Station (NGS) northeast of GCNP. National Park Service analysis of optical, air quality and meteorological data indicated a significant fraction of the haze in GCNP during this time period was due to sulfates resulting from NGS emissions (Malm *et al.*, 1989).

Salt River Project (SRP), the operators of Navajo Generating Station, conducted a study during early 1990. The SRP study also indicated a contribution of NGS emissions to haze in GCNP, but at a lower frequency of occurrence. A difference in prevailing meteorological conditions during the years of the NPS and SRP studies would at least partially account for the differences in magnitude and frequency of impacts identified by the two studies. The results and limitations of the NPS and SRP studies are described briefly in section 2.

Based on these studies and additional evidence presented, EPA has proposed regulations that would require substantial reduction of sulfur dioxide emissions from NGS. While NGS has been linked to a portion of the haze at GCNP, it is generally recognized that a number of other area and point sources also contribute to haze at GCNP. One potential source is the Mohave Power

Project (MPP), a 1580 Megawatt, coal-fired steam electric power plant located in Laughlin, Nevada, southwest of GCNP and operated by the Southern California Edison Company (SCE). Like NGS, MPP has no pollution control equipment for sulfur dioxide. Congress, desirous of additional information concerning the sources of visibility impairment in GCNP, added $2.5 million to the fiscal 1991 appropriation for EPA to conduct "a pollution tracer study at the Mohave Powerplant". Project MOHAVE (Measurement Of Haze And Visual Effects) is EPA's response to this congressional mandate.

Goals of the Study

The primary goal of Project MOHAVE is to determine the contribution of the MPP to haze at GCNP and other mandatory Class I areas where visibility is an important air quality related value. This implies a quantitative evaluation of the intensity, spatial extent, frequency, duration and perceptibility of the MPP contribution. The improvement in visibility that would result from control of MPP emissions is included in the primary goal. Secondary goals include an increased knowledge of the role of other sources on haze in GCNP and the southwestern United States in general. Because knowledge of regional transport and air quality levels is necessary to separate the effect of MPP from other sources, meeting the primary goal will result in increased knowledge about the impacts from other sources.

It is hypothesized that the maximum impacts of MPP on visibility at GCNP occur during periods with clouds present (to facilitate transformation of SO_2 to sulfate) and wind directions that transport the MPP plume toward GCNP. The study is designed to test this hypothesis.

Project MOHAVE Organization

The EPA Office of Air Quality Planning and Standards (OAQPS) in Durham, North Carolina has overall management responsibility for Project MOHAVE. Robert Bauman is the manager of Project MOHAVE and has selected staff from the Environmental Monitoring Systems Laboratory (EMSL) in Las Vegas as the technical advisors. Staff includes Marc Pitchford, a National Oceanic and Atmospheric Administration (NOAA) employee assigned to EPA and Dr. Mark Green, a Desert Research Institute (DRI) employee working under a cooperative agreement with EPA. To be advised in the overall direction of the study, Mr. Bauman has formed a steering committee composed of government and industry scientists. The steering committee includes:

Dr. Carol Ellis	Southern California Edison Company
Dr. William Malm	National Park Service
Dr. Peter Mueller	Electric Power Research Institute
Marc Pitchford	EPA (EMSL-LV)

Dr. William Wilson EPA (AREAL)

Temporary technical advisory panels provided recommendations during a planning workshop, as discussed later in this section. Coordination committees, composed of Project MOHAVE participants and their contractors responsible for various components of the study, will meet on an ad hoc basis to refine and coordinate in the following areas:

(1) Monitoring
(2) Modeling
(3) Data Management
(4) Data Analysis

These committees will facilitate joint analyses with SCE and other contributing participants. The participants in Project MOHAVE include Federal agencies, universities and private companies. A list of the main participants and their areas of responsibility is given in the summary table presented in Appendix 1.

Study Planning and Review to Date

The first significant planning effort was the formulation of a conceptual study plan. The conceptual plan outlined the main components of the study and gave generalized approaches for each aspect of the study. Preliminary monitoring locations and schedules were also identified. The purpose of the conceptual plan was to serve as a preliminary planning document to provide a common starting point for outside review. The conceptual plan was reviewed by (1) the Project MOHAVE steering committee, (2) members of the Haze in National Parks and Wilderness Areas Committee of the National Research Council, National Academy of Sciences (3) participants in a Project MOHAVE planning workshop (a group of about 40 experts), and (4) various other individuals. The conceptual plan underwent several revisions; the most recent version, which led to the current plan, is presented in Appendix 2.

The Haze in National Parks and Wilderness Areas Committee was briefed on the conceptual plan on March 14, 1991 at the University of California-Irvine. Individual members of the Committee asked clarifying questions and made some suggestions on the conceptual plan. Several of the members made additional comments at later dates. The Committee as a whole did not comment on the plan.

During the week of April 23, discussions were held between SCE, DRI, and EPA in Las Vegas to formulate conceptual models of conditions during which MPP emissions may be transported to GCNP. This included a review of the dynamic processes affecting MPP plume transport and dispersion, and the diurnal and seasonal variation of these processes. Also considered were issues concerning chemical transformation and deposition, in particular gas-phase and aqueous phase oxidation and the roles of clouds and H_2O_2. These discussions and

a summary of the meeting provided by SCE and DRI helped in selecting the intensive study periods as well as providing insight about the important physical mechanisms.

A planning workshop was held April 30-May 2 in Denver. Thirty-nine individuals with expertise in one or more study components attended. A plenary session was held first during which the conceptual plan was presented. Following the plenary session, subgroups met to make recommendations on the study components. The subgroup topic areas were: 1) tracer, 2) air quality measurements, 3) emissions, 4) deterministic modeling and upper air measurements, and 5) quality assurance. Another plenary session followed, during which clarifying questions were asked and different subgroups coordinated their plans. The subgroups again met to compile recommended study components; these were presented in a final plenary session. After the workshop, a small group met to evaluate the recommendations and plan the implementation of the study. A list of the participants attending the workshop appears in Appendix 3.

In July 1991, a table summarizing the main components of the study and the responsible persons for each component, and a map showing expected monitoring locations were prepared. These were sent out to study participants. The purposes of the summary table and map were to provide an update on the plan and to ensure that the Project MOHAVE staff and other study participants had a mutual understanding of the responsibilities and plans for each study component. The summary table was updated after review by participants. It is presented in Appendix 1. More detailed descriptions of the information in the summary table appear in subsequent sections of this plan.

Study Schedule

The field measurement portion of the study will last for one year, from September 1991 through August 1992. Intensive monitoring and tracer release periods are scheduled for January 4-31, 1992 and July 15-August 25, 1992. A list of milestones of and anticipated dates of completion major operational phase is given below. Coordination, data review, and planning meetings will be scheduled as appropriate.

MILESTONE	DATE
Deploy year-round monitoring equipment	9/91
Deploy winter intensive equipment	11/91-12/91
Winter intensive study	1/92
Begin data processing	3/92

Preliminary analysis of winter intensive	5/92
Deploy summer intensive equipment	6/92
Summer intensive study	7/92-8/92
End monitoring	9/92
Preliminary analysis of summer intensive	12/92
Receive final monitoring and modeling data	3/93
Draft report	7/93
Final report	12/93

Plan Organization

This plan is composed of 12 sections and 8 appendices. Section 2 discusses current knowledge, including recent tracer studies and data available for further study. Section 3 provides an overview of the field study design in terms of monitoring locations and schedules. Section 4 describes the tracer aspects of the study. Sections 5-7 discuss the air quality, meteorological, and optical monitoring plans. Emission inventory and source characterization are outlined in Section 8. In Section 9, data management and validation are discussed. Section 10 details the descriptive data analysis and interpretation study components. The methods of attribution to be used appear in Section 11. Section 12 describes the overall quality assurance plan.

2. Current Knowledge and Available Data

Setting

MPP is located at Laughlin, NV, about 125 km south-southeast of Las Vegas, 350 km northeast of Los Angeles, and 340 km northwest of Phoenix (see Figure 1). The MPP is a coal-fired, base loaded generating facility with a 153 m high stack. The base of the stack is at 210 m msl. It uses low sulfur (0.6% by wt.) Arizona coal delivered by slurry pipeline. Its SO_2 emission rate averages about 150 tons/day at full operation (Nelson, 1991).

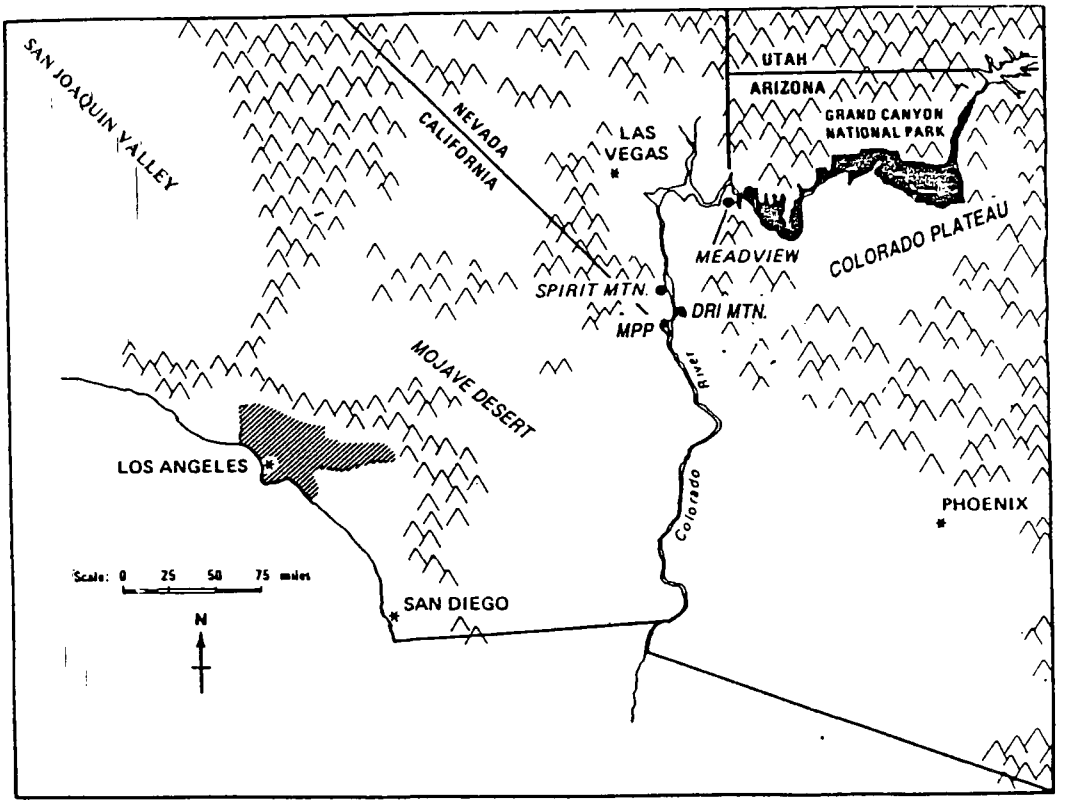


Figure 1. Map of the southwestern U.S. illustrating location of MPP.

The topography in the vicinity of the MPP is complex with sparse vegetation. A portion of the Colorado River Valley, the Mohave Valley, lies to the north of the MPP between Davis and Hoover Dams. The Mohave Valley is bordered on the west by the El Dorado and Newberry Mountains and on the east by the Black Mountains. Long north/south oriented valleys lie to the east (Detrital Valley) and west (El Dorado Valley) of these ranges.

The Mohave Valley walls are not symmetric with respect to the valley axis. Western slopes rise gradually, while eastern slopes rise slowly for the first few kilometers with steep walls further to the east. The border between Nevada and Arizona also extends along the valley axis. The bottom of the valley is about

200-300 m msl and the ridges reach 1200 m msl. Toward the west, the Mohave Valley extends into a high plateau and toward the east into the Detrital Valley plateau (600 m msl). The Mohave Valley narrows significantly as it approaches Hoover Dam. At Lake Mead the terrain flattens. The western entrance to GCNP is at the end of the eastern arm of Lake Mead (180 m msl).

This terrain controls the mesoscale, but not the synoptic scale flow patterns.

Transport Regimes

Several modeling and measurement studies have been conducted in the vicinity of the MPP over the past 20 years (Freeman and Egami, 1988; Yamada, 1988; Koracin *et al.*, 1989; White *et al.*, 1989). Results from these studies provide a conceptual model of pathways by which MPP emissions can reach GCNP. Figure 2 illustrates the three synoptic flow patterns of greatest importance; (1) summertime dry southwesterly flow (flow from the southwest toward the northeast), (2) summertime monsoons, and (3) winter storms.

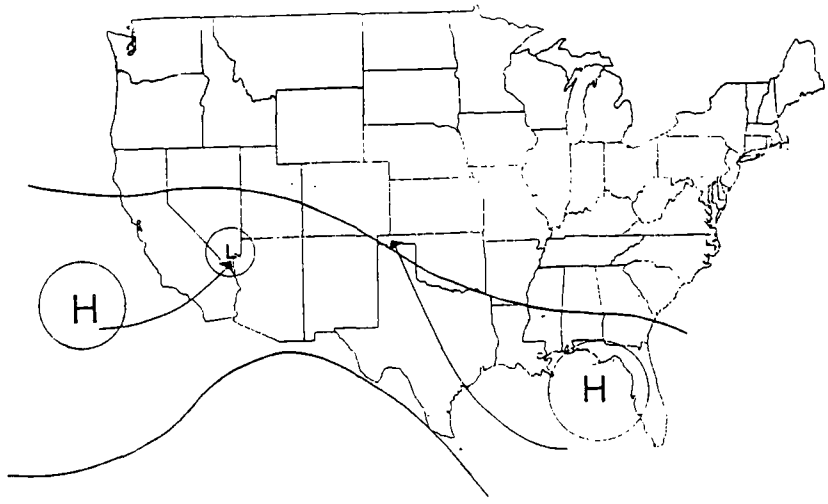
Both mesoscale and synoptic scale meteorological conditions influence the movement of the MPP plume. The relative influence from each of these transport and transformation scales differs from summer (June, July, August) to winter (December, January, February). Southerly to southwesterly flows are needed to transport MPP emissions to the GCNP. Spring and fall are transitional periods that contain mixtures of the summer and winter regimes and are not as well-differentiated from each other. Figure 3 illustrates the dominant air flow for each quarter of 1990 as derived from the Dri Mountain wind data.

During the summer, southwesterly, westerly and southerly winds are common in the vicinity of the MPP. There are two distinct cases; one with dry airmasses and a second with moist monsoon airmasses, respectively. During the winter, the most common situation is northerly winds associated with a high pressure ridge over the Pacific Coast. However, infrequent frontal passages result in westerly and southwesterly flows on the order of 10% of winter days. The latter conditions can transport MPP emissions toward GCNP.

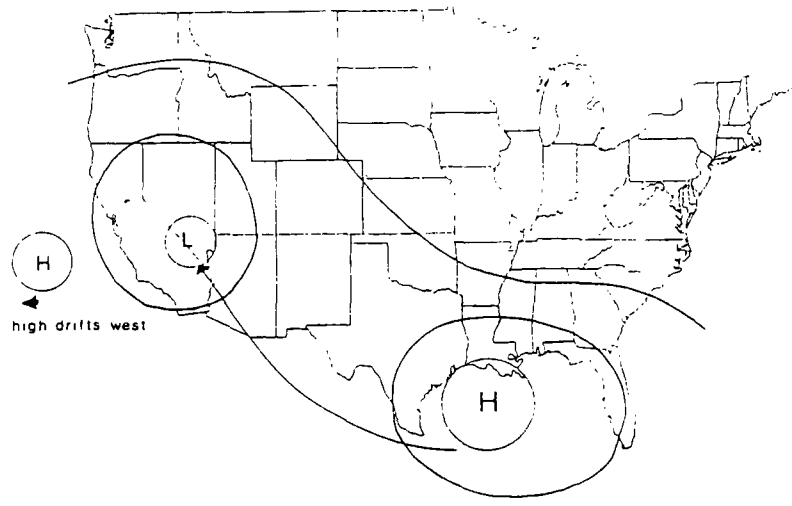
Dry, Southwesterly Flow from Southern California and the Pacific Ocean

The most common occurrence is dry, southwesterly synoptic flow caused by heating of the Mojave Desert which creates a lower pressure with respect to incoming air masses. These air masses traverse the Mojave Desert after entraining pollutants emitted from urban southern California. These include pollutants flowing through Tehachapi, Cajon and Banning passes.

This scenario has a high frequency of occurrence during the summer months. The regimes change daily from decoupled flow during the night with localized circulation patterns within the Mohave Valley and along the slopes of



Summer Monsoonal Flow



Winter Storms

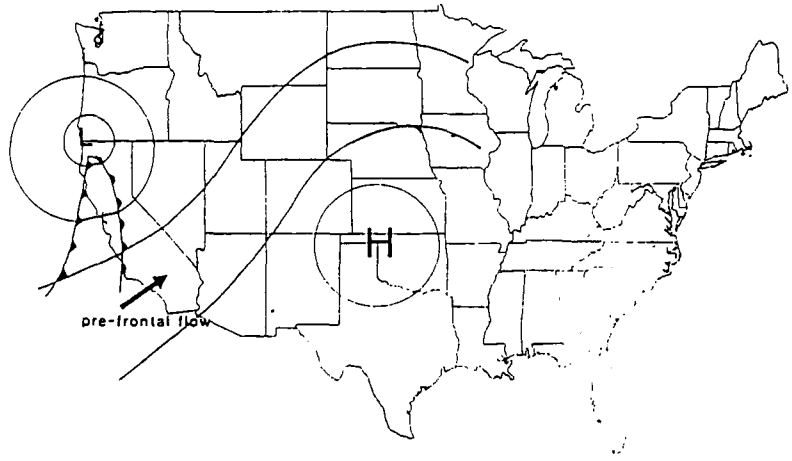


Figure 2. Synoptic flow patterns of concern. a) Dry summer southwesterly flow. b) Summer monsoonal flow. c) Winter storms.

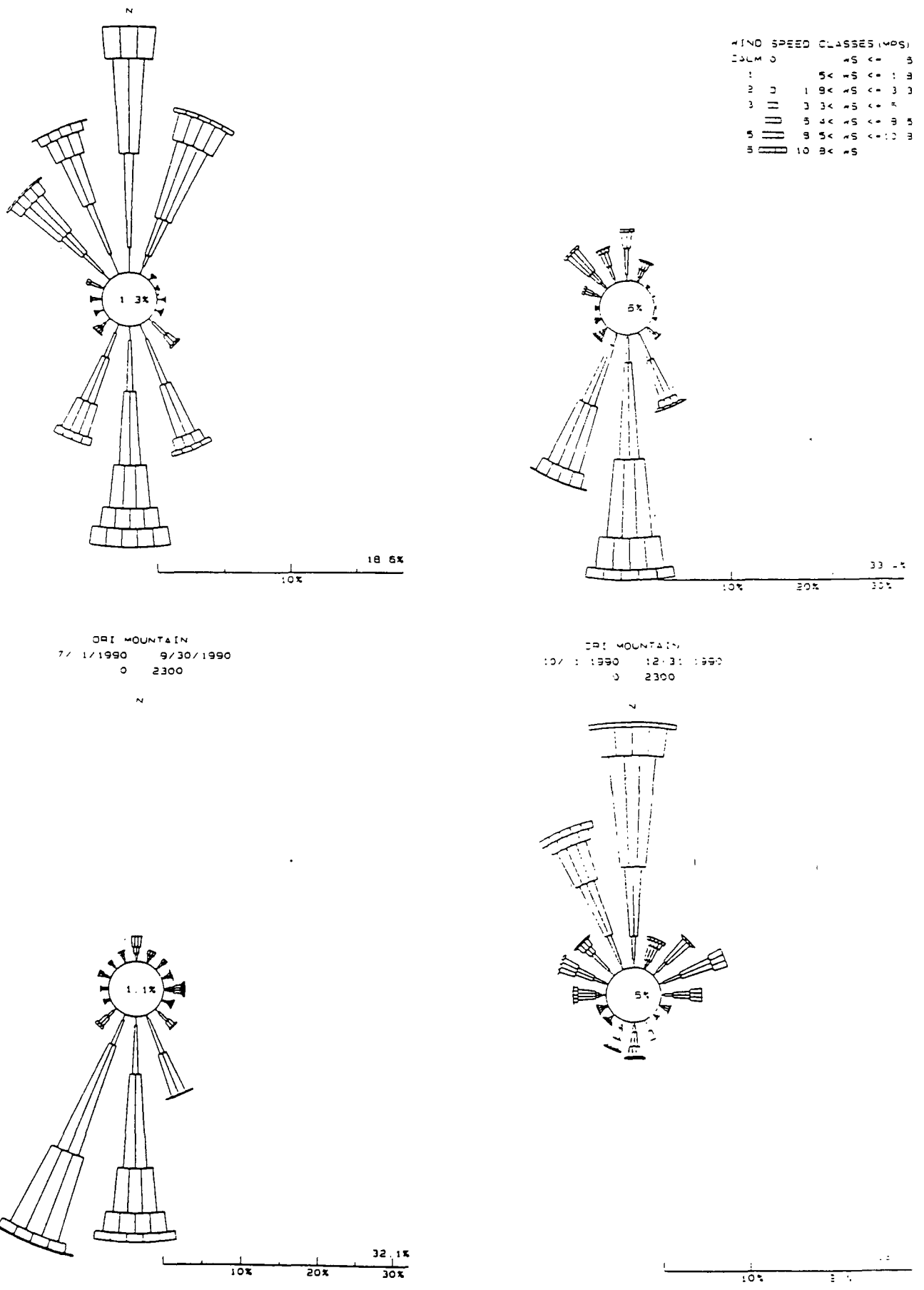


Figure 3: Dri Mountain quarterly wind roses from 1990.

the constraining mountains, to coupled flow that is dominated by the synoptic winds aloft.

Summer Monsoons

During July and August, moist air is frequently transported from the Gulf of California and/or the Gulf of Mexico in southeasterly to southerly flows. Synoptic wind speeds vary from 6 to 20 m/s at 6000 m agl. These air masses traverse northern Mexico, the southern part of Texas, New Mexico, and most of the state of Arizona. Pollutants emitted from the smelters in Arizona and Mexico as well as those from Phoenix and Tucson can be entrained into this airmass. This synoptic flow is driven by a large-scale low over the western part of the U.S. created by strong surface heating.

Differential heating causes updraft motions on the slopes of mountain on both sides of the valley, resulting in chains of clouds developing along the ridgetops. These clouds may offer a mechanism for rapid oxidation of SO_2 to SO_4 if the plume is entrained in them and if oxidants such as H_2O_2 are present in sufficient amounts.

The reacted and unreacted emissions could then be carried through the Mohave Valley by the southerly component of the wind toward Lake Mead, after which they might be transported toward GCNP by locally channeled flow or caught up in the monsoonal flow and transported across the plateau to the GCNP. Summer monsoon episodes are usually of 3 to 5 days in duration.

Winter Storms

In general, the synoptic weather patterns are not as favorable for transport from MPP towards GCNP in winter. The Great Basin and the Colorado Plateau are frequently dominated by high pressure cells creating a flow that is not conducive for transport from MPP to GCNP. Southwesterly to westerly flow occurs mainly during the movement of frontal systems, developing over the Pacific Ocean from west to east. These storms in general exhibit a minimal warm frontal activity. As a consequence the southwesterly to westerly flow needed for transport from MPP to GCNP will occur as the cold front with its associated trough approaches the Mohave Valley. This weather type can last from a day to three days, be wet or dry, and usually there are about ten cases during the winter period.

WHITEX

The Winter Haze Intensive Tracer Experiment (WHITEX), conducted by the National Park Service, was designed to evaluate the feasibility of attributing single point source emissions to visibility impairment in selected geographical regions. WHITEX was conducted during a six week period in January and

February 1987. During this time, an artificial tracer, deuterated methane (CD_4), was released from the NGS. Aerosol, optical, tracer and other properties were measured at Hopi Point, which is in GCNP, and other locations. Synoptic weather maps indicated a high frequency of high pressure over the area, which resulted in transport of the NGS plume from the northeast toward GCNP. Trajectory analysis and deterministic modeling indicated transport from the area of NGS to Hopi Point during the period with highest sulfate concentrations.

The extinction budget at Hopi Point indicated that sulfate aerosol (and associated water) contributed two-thirds of the non-Rayleigh light extinction during WHITEX. Attribution analysis used the Tracer Mass Balance Regression (TMBR) receptor model and the Differential Mass Balance (DMB) hybrid model. According to the NPS analyses, NGS contributed substantially to sulfate and light extinction at Hopi Point.

The WHITEX data analysis methodology, results, and use of the results were cause for considerable controversy. The Committee on Haze in National Parks and Wilderness Areas evaluated WHITEX (National Research Council, 1990). The Committee neither fully supported or fully discredited the WHITEX report. Based on evaluations of meteorological, photographic, chemical and other physical evidence, the Committee concluded "at some times during the study period, NGS contributed significantly to haze in GCNP." However, the committee also concluded that "WHITEX did not quantitatively determine the fraction of $SO_4^=$ aerosol and resultant haze in GCNP that is attributable to NGS emissions."

A key uncertainty identified by the Committee is the use of TMBR and DMB to apportion secondary species such as $SO_4^=$. Limitations of the regression analyses noted by the committee are: "(1) satisfactory tracers were not available for all major sources; (2) the interpretation did not adequately account for the possible covariance between NGS contributions and those from other coal-fired power plants in the region; and (3) both models employ inadequate treatment of sulfur conversion, which is an important controlling factor in the formation of haze at GCNP." Another limitation noted by the Committee was the lack of measurements within the canyon (beneath the rim). A more complete review of the National Research Council WHITEX evaluation is provided in Appendix 8.

SRP Study

The Navajo Generating Station Visibility Study was conducted for the SRP, the operators of NGS, from January 10 through March 31, 1990. Its purpose was to address visibility impairment in GCNP during the winter months and the level of improvement that might be achieved if SO_2 emissions from NGS were reduced. The study was performed to provide input to the rulemaking process of the EPA regarding NGS SO_2 controls (Richards *et al.*, 1990).

Perfluorocarbon tracers were released from each of the three stacks of NGS. Surface and upper air meteorology, particle and gaseous components, and

tracer measurements were made at many sites. Deterministic modeling was done to estimate the contribution of NGS and other sources to sulfate levels for two 6 day periods with poor visibility. Various data analysis techniques were used to examine the relationships among NGS emissions, meteorology, air quality, and visibility during both episode and non-episode conditions.

The SRP study concluded that NGS emittants were absent from the vicinity of Hopi Point most of the time . The study estimated that the average contribution of NGS to fine sulfur at Hopi Point was small, although NGS sulfur dominated during one 4-hour period. However, it was noted that the frequency of wind directions transporting the plume toward GCNP were lower than normal during this time period.

MPP Emission Modulation Studies

The MPP was inoperable for the seven month period June through December 1985. Using data from the period of shutdown and during operation of MPP, a study was done by SCE (Murray *et al.*, 1990) to assess the effect of MPP upon particulate sulfur levels at Spirit Mountain, Meadview, and Hopi Point. Spirit Mtn. is 20 km northwest of MPP, Meadview 110 km north-northeast of MPP and Hopi Point 240 km northeast of MPP. Meadview, 5 km west of the boundary of GCNP, is expected to have the highest particulate sulfur impact from MPP among the three sites. The study found no statistically significant difference in sulfate levels at the three sites between operation and shutdown of MPP. It was suggested that the substantial year to year variability of sulfate was responsible for not detecting a statistically significant difference. The 95% confidence bounds for the MPP impact was from less than 11.6% to less than 21% at Meadview and less than 3.3% to less than 7.8% at Hopi Point during favorable transport conditions. The upper limit on average sulfate at Meadview was estimated to be 15%, which is the level of uncertainty in the statistical analysis.

From data presented by Murray, it can be seen that sulfate levels at Spirit Mountain, generally not affected by MPP, were greater during the outage compared to non-outage periods, indicating higher background levels during the outage. However, at Meadview, average sulfate levels were lower during the outage. Thus, levels at Meadview were lower during the outage even though regional levels were higher. While suggestive, the number of samples was not sufficient to prove an impact from MPP. This comparison, done as part of the scoping process, appears in Appendix 4.

A more sophisticated study of the outage will be conducted under the Desert and Intermountain Air Transport program at DRI, sponsored by SCE. Chemical and physical analysis of filters for the SCENES program, used in Murray's study, were analyzed only every third day. Samples for the intermediate days were archived. The new study will analyze all samples, including those previously analyzed. A more sophisticated meteorological

classification scheme will also be done. The sulfate levels for the same meteorological regimes can then be compared for the outage and non-outage conditions. Other emission modulations of shorter duration (i.e. periods where only one of the two units at MPP was operating) will also be analyzed. Deterministic wind field and transport modeling will be done for each of the meteorological regimes. The modeling will account for variations within each regime. A detailed compilation of regional SO_2 emissions for the control and outage periods will be done. A draft version of the outage study plan appears in Appendix 5.

3. General Field Study Design

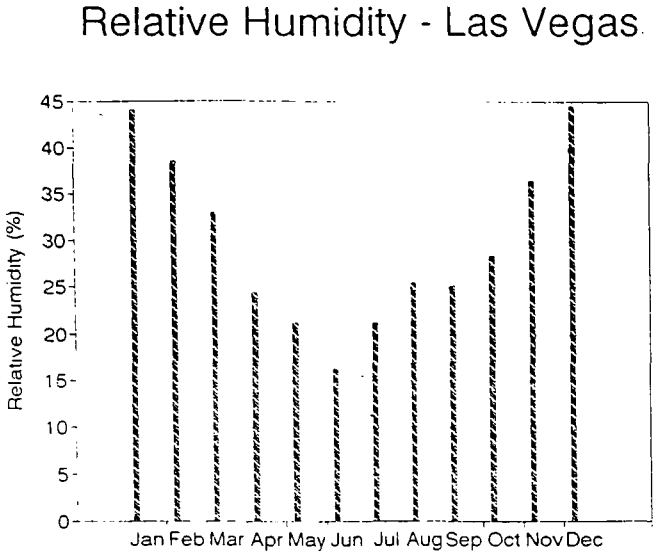
The duration of the field study will be one year. It was considered important in evaluating the overall impact of a source to consider a complete annual cycle. By monitoring for an entire year, all the seasons may be studied. For practical reasons, the year was divided into "intensive" and "non-intensive" periods. During the intensive periods tracer will be emitted from the MPP stack and tracer and particulate data will be collected continuously at over 30 sites. During the non-intensive periods tracer will not be released, the number of particulate monitoring sites will be scaled back considerably and sampling will be done only two days per week. Meteorological and optical monitoring will be done continuously.

Selection of the Intensive Periods

In selecting the intensive study periods, it was desired to select periods in which the MPP may be most likely to contribute to haze in GCNP. It is expected that secondary sulfates formed from oxidation of MPP SO_2 emissions is the largest portion of the MPP contribution to haze in GCNP. Primary particulate emissions from MPP contribute to haze nearer to the power plant, but at the distance of the GCNP, secondary sulfates are expected to dominate. Dry phase oxidation of SO_2 is much slower than aqueous phase oxidation. Thus, cloudy periods can cause much more rapid conversion of SO_2 to sulfate. Aqueous phase oxidation is on the order of 50-100% per hour if oxidants are present in sufficient quantity (Lee, 1986).

Cloudy periods with wind directions transporting the MPP plume toward GCNP are the periods when impacts to visibility at GCNP due to MPP would be most likely to occur. As discussed in Section 2, these conditions may occur during the summer monsoon and certain winter periods. Calculations of the potential impact of MPP to haze at GCNP under highly simplified conditions were done for dry southwesterly and monsoonal summer conditions, and pre-frontal winter conditions. These calculations indicated a potential for perceptible visibility impairment at GCNP from MPP emissions for all three cases (see Appendix 4).

Moisture parameters calculated from long term National Weather Service data from Las Vegas are shown in Figure 4. Specific humidity, which gives the amount of water vapor in the air, is highest in August, with July having slightly less moisture. Average monthly dew point temperature for the years 1982-1990 (Figure 5) at Dri Mountain also showed a peak in August, with slightly lower values in July. Relative humidity peaks in December and January. Also note that August has higher relative humidity than July. December and January are the cloudiest months, with February and March only slightly less cloudy. A secondary peak in cloudiness occurs in July, with somewhat less cloudiness in



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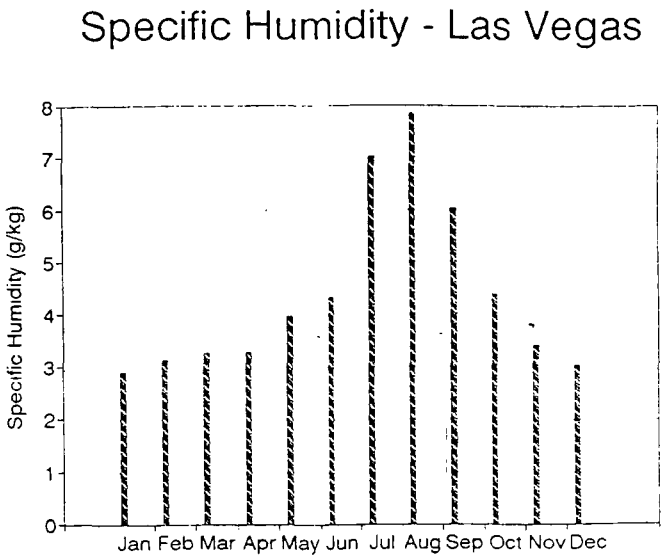

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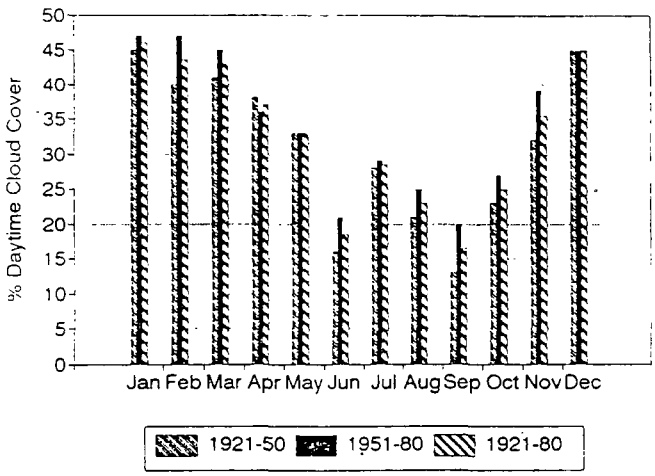

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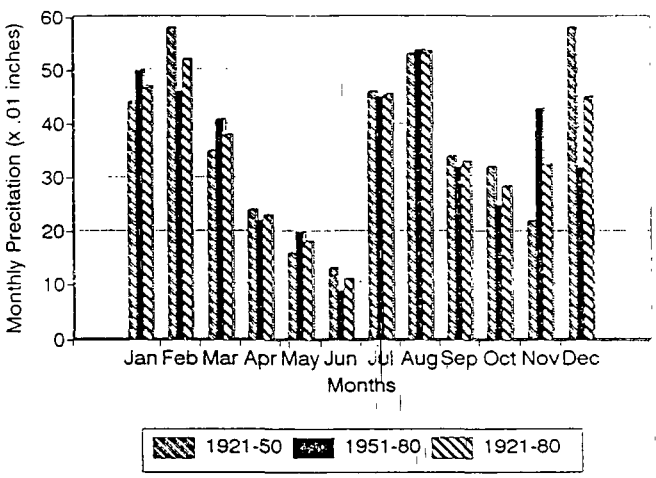

(d)

Figure 4. Las Vegas moisture climatology

(a) Average specific humidity by month: 1951-1980.

(b) Average relative humidity by month: 1951-1980.

(c) Average percent daytime cloud cover by month.

(d) Average precipitation by month.

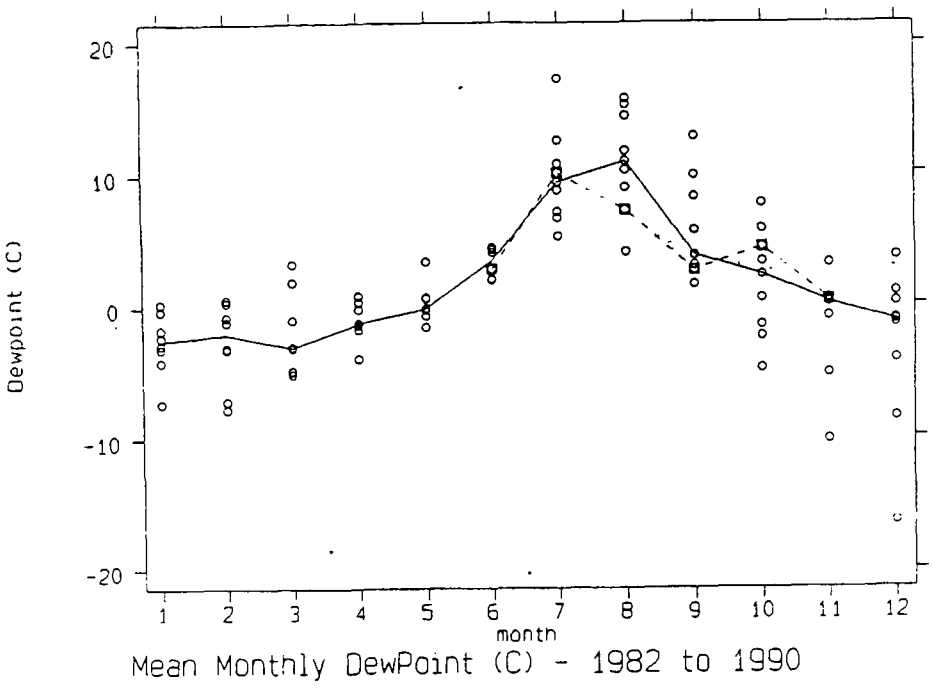

Figure 5. Mean monthly dew point temperature at Dri Mountain: 1982-1990.

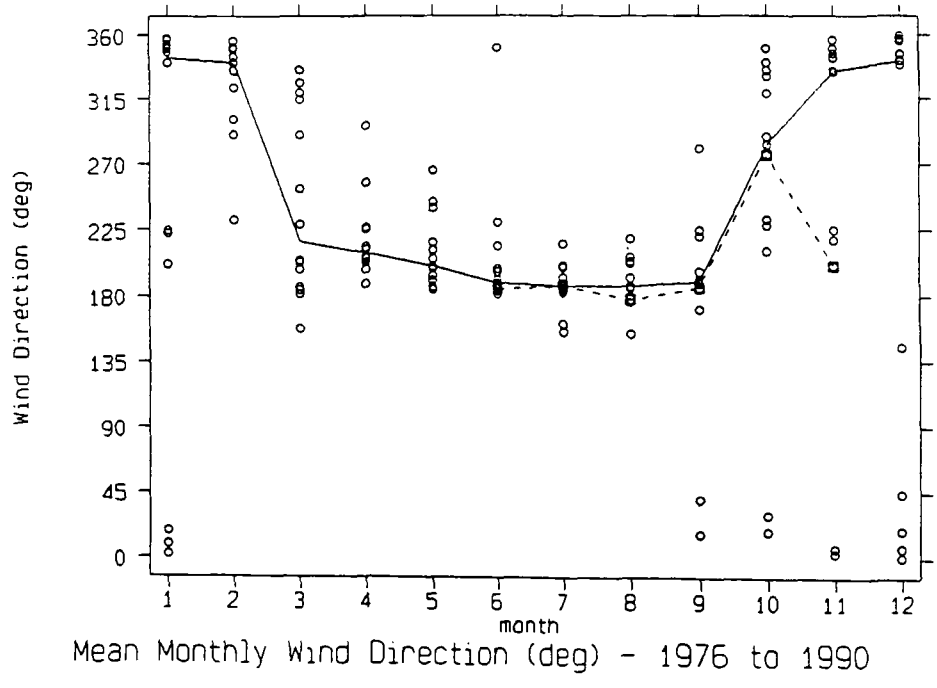

Figure 6. Mean (resultant) wind direction by month at Dri Mountain: 1976-1990.

August. The precipitation data show two distinct peaks; one is July and August, the other December through February. Of interest is the substantial difference in average precipitation in November and December between the 1921-1950 and 1951-1980 data. This suggests that year to year variability is large.

The climatology described above suggests a summer intensive period covering portions of July and August and a winter period that could be any time between December and February. January and December showed the highest values for the moisture related parameters, with January's precipitation data being more consistent than December's. January was chosen for the winter intensive. August has slightly higher relative and specific humidity than July. Thus, the summer intensive will be centered on early August.

Even though we are attempting to optimize the study periods for the specific conditions described above, meteorological conditions are highly variable from year to year. Most frequent winter flow at MPP is away from GCNP. However, winter flow is often toward the Joshua Tree Wilderness, another Class I visibility protected area. In summer, we are likely to experience dry flow from the southwest a significant portion of the time in addition to the moist monsoonal flow. Thus, information about other common conditions will also be obtained.

Mean vector (resultant) wind direction each month for the years 1976-1990 at Dri Mountain is shown in Figure 6. Dri Mountain is a pointed hill 150 meters high and adjacent to the Colorado River a few kilometers north of MPP. The instrument level is approximately at the same elevation (MSL) as the top of the MPP stack. However, the plume centerline is generally 400-700 m, averaging 663 m above stack base, which is 250-550 m above Dri Mountain. The winds at Dri Mountain would be expected to be influenced more by channeling due to topographic features than the winds at plume height, particularly during nighttime and morning hours. Winds at plume height typically have a greater westerly component (toward GCNP) than Dri Mountain winds. The winds at Dri Mountain indicate a predominance of northerly winds during November through February and southerly winds during April through September. March and October are transitional periods. Three January periods during 1976-1990 had south to southwest resultant winds, indicating more frequent flow toward GCNP during these years.

The analysis of humidity, clouds, precipitation, and winds suggest optimal of January 4-31, 1992 for the winter intensive and July 15 to August 25, 1992 for the summer intensive.

Siting of Monitoring Instrumentation

The aerosol, tracer and optical monitoring network includes three classes of sites. These are denoted as (1) receptor, (2) other Class I, and (3) background sites. A more detailed description of air quality and meteorological monitoring is described in sections 5 and 6, respectively. The aerosol and tracer monitoring was designed to provide sampling and analysis every day for many sites during

the intensive periods, and sampling and analysis two days a week during the rest of the study year. The reduction of monitoring for the non-intensive periods is necessary due to cost considerations.

The preliminary network of sites is shown in Figure 7. The siting will be finalized after a monitoring planning meeting to be held in Las Vegas in early October. A listing of the sites, approximate elevation, instrumentation, and a brief reason for selecting each site is given in the table below. The receptor sites (R1-R4) are either within or in very close proximity to GCNP. The other Class I sites (I1-I6) are in areas that may be impacted by MPP and/or serve as background sites. Most of the receptor and other Class I sites had some degree of existing or planned monitoring prior to Project MOHAVE. These sites will have supplemental monitoring associated with Project MOHAVE and will operate during the entire study year. The background sites (B1-B21) are intended to characterize high elevation and low elevation transport into the study area as well as showing more detailed concentration patterns within the study area. The background sites will operate only during the intensive periods. The instrumentation to be used is described in Sections 4-7 and references cited in those sections.

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SITE IDENTIFICATION TABLE

Id No	Name	Elevation (meters)	Particle & Tracer	Optical	Meteorology
R1	Meadview	900	1	T,N,P	S,U
R2	Long Mesa	1830	4	N	S
R3	Hopi Point	2160	1	T,N,P	S
R4	Indian Gardens	1220	1	T,N,P	S
I1	San Gorgonio	1680	2	T,P	
I2	Joshua Tree	1500	2	P	
I3	Tonto	730	2	T,P	S
I4	Sycamore Canyon	2000	2	P	
I5	Petrified Forest	1680	2	T,P	S
I6	Bryce Canyon	2600	2	P	
B1	Tehachapi Pass	1240	3		
B2	Cajon Pass	1380	3		
B3	Baker	280	3		
B4	Amboy	190	3		
B5	Parker	130	3		

B6	Wickenburg	620	3	
B7	Las Vegas Wash	370	3	
B8	Cottonwood Cove	210	3	S,U
B9	Yucca	580	3	
B10	Dolan Springs	850	3	
B11	Truxton	1370	3	S,U
B12	Seligman	1620	3	
B13	Prescott (airport)	1620	3	
B14	Overton Beach	370	3	
B15	New Harmony	1520	3	
B16	Marble Canyon	1220	3	
B17	Mt Springs Summit	1680	3	
B18	Spirit Mountain	1700	3	
B19	Hualapai Mt Park	1980	3	
B20	Camp Wood	1980	3	
B21	Jacob Lake	2400	3	

Explanatory notes:

1 Full IMPROVE samplers. 24-hour samples midnight to midnight, Wednesday and Saturday during the non-intensive periods. Twice daily samples of aerosol and tracer will be taken each day during the intensives. Specific hours for the beginning and end of each daily sampling period will be determined at the monitoring coordination meeting. DRUM samplers with 4 or 6 hour sampling periods (only selected samples will be analyzed).

2 Full IMPROVE samplers. 24-hour samples (aerosol and tracer) each day during the intensives. 24-hour samples midnight to midnight Wednesday and Saturday during non-intensive periods.

3 IMPROVE channel A and filter pack for SO_2. 24-hour samples (aerosol and tracer) each day during the intensives. No sampling during non-intensives.

4 Long Mesa will only have a DRUM sampler for particle monitoring.

T= transmissometer, N= nephelometer, P= photography, S= surface meteorology, U= upper air meteorology

Surface and upper air meteorological data will be collected at additional sites identified in Section 6.

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Important considerations in selecting sites include the availability of power and accessibility. The power requirement imposes strict limitations on siting.

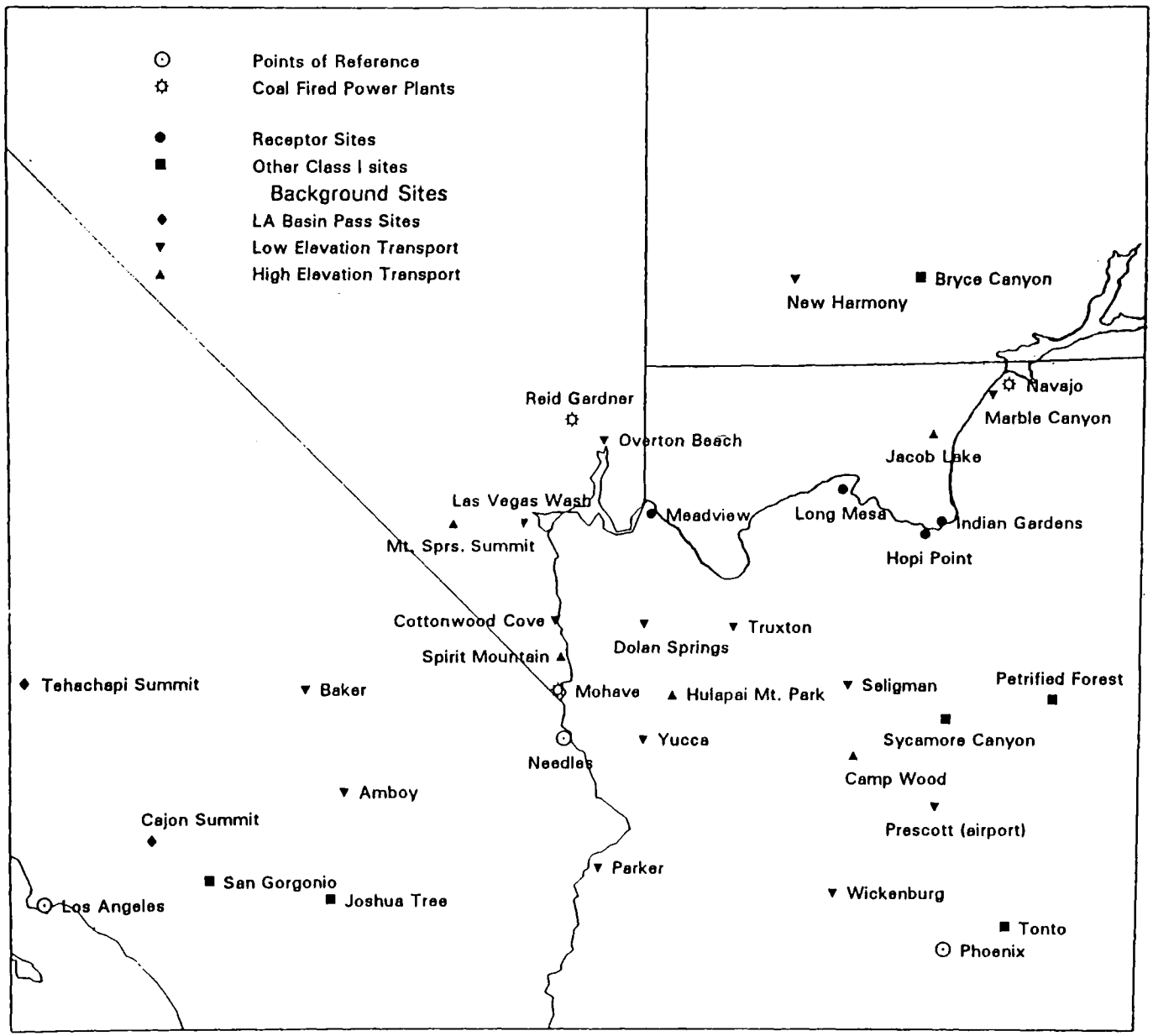

Figure 7. Location of air quality and tracer monitoring sites.

Meadview was chosen because it is within about 5 km of GCNP, has existing monitoring by the Desert Research Institute (DRI), and is at the west end of GCNP, thus closer to MPP than other areas of GCNP. Long Mesa is also at the edge of GCNP and the location of another DRI monitoring site. Hopi Point and Indian Gardens are existing NPS monitoring sites within GCNP. Joshua Tree and Sycamore Canyon are potentially impacted by MPP. The remaining Class I sites will help characterize transport into the area.

The Tehachapi Pass site (B1) is located in a pass between the San Joaquin Valley and Mojave Desert and is intended to monitor the exchange of air between these areas. The San Joaquin Valley is a large source of SO_2. The Cajon Pass site (B2) is located between the Los Angeles Basin and Mojave Desert and is a major exit pathway for Los Angeles Basin air. Sites B3-B6, I2, and I3 are low elevation southern boundary sites. These locations form an arc to characterize the sulfur flow into the main study area from the southwest to southeast.

Locations B17-B20 form a second arc to the south of GCNP. These sites are located on terrain that rises 900 to 1200 meters above the surrounding area. The sites should be in the middle of the mixed layer during the summer intensive and frequently above the mixing layer during the winter intensive. Measurements from these sites when tracer is absent, coupled with the nearby low-elevation southern sites B7-13, should characterize the sulfur flux from the southwest through southeast exclusive of MPP sulfur into the receptor area. At other times, tracer from MPP may be present at these sites. In conjunction with the low elevation southern sites, these sites will help determine vertical distributions of sulfur and tracer.

Sites B7-B13 are located in possible MPP transport corridors between the southern boundary sites and GCNP. These locations will indicate if the emissions from MPP are transported toward GCNP in a narrow cone or more widely dispersed air mass, in addition to identifying the most common transport corridor from MPP to GCNP.

The MPP plume usually travels to the south along the Colorado River in the winter. It is suspected that the plume may sometimes leave the river area in an eastward direction through a gap in the mountains near site B9. The high elevation sites B18-B20 along with B10-B12 should be able to verify if MPP emissions are being transported from the area of B9 to the east or northeast in a low-level surface layer or more dispersed in a deeper layer.

Locations B7, B8, B10, and B14 are placed in an attempt to isolate emissions of MPP, the Reid Gardner power plant, and Las Vegas as they are mixed over Lake Mead on their way to the western end of GCNP (Meadview). Under stagnation conditions, the high elevation sites B17 and B18 should characterize the cleaner regional air above the mixing layer.

The northern boundary sites B15, B16, B21, and I6 are located to characterize flow into the region from the north. These sites will help identify the effects of the Wasatch Front urban and industrial sources, the NGS, and other coal-fired powerplants to the north and east. Site B15 is very close to Zion

National Park. Flow from MPP is likely to be transported toward this site often during the summer. Locations B15 and B16 serve as low elevation sites. I6 and B21 are high elevation sites.

4. Tracer

Purpose

During the intensive study periods, an artificial tracer will be released from the stack of the MPP and monitored at the same 31 locations as the air quality monitoring. There are several reasons for releasing tracer. Tracer monitoring data will identify the general transport patterns for the MPP plume. Knowing where the plume goes is critical to begin to understand the larger question of the MPP's visibility effects. To fully resolve the plume position and extent, a very extensive monitoring network, including aircraft measurements would be required. This is beyond the resources of the study. The 31 monitoring locations should provide the approximate location of the plume, although its horizontal and vertical extent will be uncertain.

Different artificial tracers will be released from the Los Angeles Basin and San Joaquin Valley during one-half or more of the summer intensive. These additional tracers will be released to gain insight into the transport of emissions from these large source areas into the Project MOHAVE study area. They will help identify the interaction between MPP and southern California emissions and provide dilution ratios for southern California emittants.

The tracer will be used to provide a check of deterministic modeling results. A transport model, using wind fields generated by a dynamic meteorological model, will predict plume locations. The tracer data will be compared to the transport model predictions to evaluate the model performance. The concentrations of the tracer will be used to evaluate dispersion of the plume predicted by the models as well as location. The dynamic meteorological and transport models are discussed in section 10.

The tracer will also be used for receptor and hybrid modeling purposes. The tracer will serve as a unique "tag" for the MPP. The receptor and hybrid modeling is described in section 13.

Choice of Tracer

Ideally, a tracer should closely mimic the species of interest for receptor modeling and chemical transformations; in this instance SO_2 and its conversion to SO_4, and deposition of the sulfate particles. This would suggest using isotopes of sulfur or oxygen. However, the large amounts of these materials that would be required are not available; to produce them would require resources greater than those available for this study.

Among the potential tracer materials are deuterated methane (CD_4), various perfluorocarbons (PFT's), Sulfur hexafluoride (SF_6), and particulate rare earth oxides. CD_4 and PFT's and SF_6 are conservative tracers; thus conversion of SO_2 to SO_4 and deposition of SO_2 and SO_4 can not be directly simulated. It has been suggested that nonconservative rare earth particle tracers be used

because of their potential to mimic sulfate particles. However, sulfate particles are not directly emitted in significant quantities; rather they are typically formed during transport at rates which vary with meteorologic and other atmospheric conditions. Thus some variable proportion of the rare earth particles will have deposited before the sulfates are formed. Additionally the deposition of SO_2 occurs more rapidly than either sulfate or rare earth particles. A combination of conservative and particulate tracers could yield additional insights into the fate of MPP emissions than that obtained using a single tracer. However, the additional expenses associated with using an additional class of tracer is beyond the resources of Project MOHAVE.

SF_6 has been used in many short range experiments. Although the cost per kilogram is low compared to other conservative gaseous tracers, the background concentration is much higher, which more than offsets the decreased unit cost. SF_6 is not practical for the spatial scale of the study region.

CD_4, used in WHITEX has low background values and is detectable at very low concentrations, so small amounts of this tracer are sufficient. Though the cost per unit mass is high, the total cost of tracer material is less than the cost of PFT's. However, the sample analysis cost is very high ($800-$1000/sample), compared to about $20/sample for PFT's. The lower cost of PFT analysis encourages the analysis of many more samples for the available budget. For CD_4 the strategy is to analyze a subset of all possible samples. Analysis of all samples allows a more thorough evaluation of the deterministic modeling. Different PFTs can be released from other sources of interest and analyzed from the same sample for virtually the same low analytical cost.

The SRP tracer study, which used PFT's, apparently had some major problems with the tracer portion of the study. Collocated samplers showed near zero correlation. Apparently this was at least partially due to the fact that many samples were near the limit of detection. Other tracer studies have also had apparent quality control problems, for example, contaminated samples. It is imperative to have a rigorous quality control program for the tracer components of the study. The quality control methods to be used for the Project MOHAVE tracer study is described later in this section. There is no fundamental reason that would prohibit PFT's or other tracers from giving reliable, quantitative results.

Project MOHAVE will use perfluorocarbon tracers. The tracer to be used to track the MPP plume is ortho-cis perflorodimethylcyclohexane (ocPDCH). The tracer material to be released is ortho (o) PDCH, 45% of which is ocPDCH. Perfluoromethylcyclopentane (PMCP) will be used to tag the Los Angeles Basin. Perfluorotrimethylcyclohexane (PTCH) will be used to track emissions from the San Joaquin Valley. The ambient background of ortho-cis PDCH is very low, 0.3 parts per quadrillion (ppq) (Dietz, 1987). The SRP study used PDCH and other PFT's but analyzed for total PDCH, not individual isomers. The background of total PDCH is 22 ppq. PMCP background concentrations are 3.3 ppq; PTCH background is 0.3 ppq. Brookhaven National Laboratory (BNL) will

do the tracer analysis for Project MOHAVE. In addition to analyzing isomers, BNL pre-concentrates the sample; thus much greater sensitivity is achieved compared to the SRP analysis methodology (Dietz, 1991).

Tracer Release

Tracer can be released at a constant emission rate or at a constant ratio of tracer to SO_2. Variation of tracer to SO_2 ratios was a complicating factor in the WHITEX receptor modeling analysis. If released at a constant rate, SO_2 emission rate variations would complicate the receptor modeling, requiring adjustment of the ratio of tracer to sulfur dioxide concentration. This requires knowledge of plume age. However, for use in deterministic modeling, it is more desirable to have a constant tracer emission rate, to simplify the dispersion calculations. If a constant release rate were used, the deterministic model would be used to give the plume age necessary to adjust the tracer to sulfur dioxide emission rates in the receptor modeling. The MPP is a base loaded unit. It typically operates at either full capacity, 1/2 capacity (one unit down) or is down. Tracer will be released at a rate proportional to the SO_2 emissions if a practical approach to do it can be devised. If not, then the tracer release rate will track the status of the power generation units with full, one-half or zero tracer emissions, corresponding to two, one, or zero units operating. This will more closely preserve the ratio of tracer to SO_2 emissions than a constant tracer release rate. Good coordination between MPP operators and the tracer release personnel will be expected. Tracer release from the San Joaquin Valley and Los Angeles Basin will be at a constant rate.

Release Equipment

The perfluorocarbon tracer liquids are very similar in viscosity to silicone fluids, but are quite dense (densities from 1.7 to 1.8 g/mL liq.). Large release rates, tens of kilograms per hour, have been accomplished with (1) atomizers spraying directly into the air, or (2) by vaporizing a PFT liquid stream, diluting with air below the PFT dewpoint at the exit, and emitting the diluted stream into the air or other fluid (such as the flue gases going up a power plant stack).

For low release rates, tenths of kilograms per hour, such as will be needed for Project MOHAVE and as was used in METREX in 1984 (Draxler, 1985), the tracer can be released by evaporation using the METREX-designed equipment. The release unit has only two moving parts: a squirrel cage fan motor and a metering pump. The tracer flows in a closed circuit from the reservoir through the peristaltic pump rollers (the tubing is compressed to move the liquid) directly into the airstream on the surface of a heated disk. The disk and heater are located in a cylindrical mixing chamber. The heater, adjustable up to 600 W, maintains the temperature of the disk above the tracer's boiling point. The

system's electronics control the duration of release and the duration that the system is off. Times for each on-off cycle can be set by tens/whole/tenths of an hour for each cycle. A small strip chart recorder notes when the pump and heater are on. The pump rate is preset on a calibrated dial. The airflow should be sufficient to ensure all the vapor is diluted below the saturation mixing ratio for the expected ambient temperature without blowing the tracer drops off the heater before they vaporize.

Three release units were built by the NOAA Air Resources Laboratory in Silver Spring, MD, and now reside at their laboratory in Idaho Falls. The system was designed to handle release rates of the magnitude needed for Project MOHAVE. However, substantial design changes may be made by NOAA in consultation with Brookhaven in order to insure reliable operation (including accurate release rates and constancy of release).

PFT Programmable Samplers

Each site will be equipped with a programmable Brookhaven atmospheric tracer sampler (BATS). The sampler was initially developed by BNL and was commercially manufactured by the Gilian Instrument Corporation (West Caldwell, New Jersey). The unit consists of two sections: the lid, containing the sample tubes, and the base, containing the power control. The entire unit is housed in a weather-resistant 36 cm x 25 cm x 20 cm container and weighs approximately 7 kg. Power is supplied by an internal rechargeable nominal 8-VDC battery for operation at remote locations, or by a charger where 155-VAC is available. For Project MOHAVE, each unit must be run on a charger in order to collect the full twenty-three (23) 36-or 72-L air samples.

The BATS removable lid holds 23 stainless steel sampling tubes, each packed with approximately 150 mg. of Ambersorb adsorbent. The Ambersorb adsorbs the tracers from the sample air flowing through the tube. Breakthrough of the perfluorocarbon tracer gases is less than 0.1%. The tracer gases remain adsorbed until extreme heat is applied to the tube to desorb the tracer at analysis time. The sample air flow is directed consecutively through the adsorbent tubes by means of a multiple port switching valve which is controlled by the base. Since the lid is removable and interchangeable, multiple lids can be used on a single base.

The BATS base contains a DOE-Environmental Measurement Laboratory constant mass flow pumping system (Latner, 1986) which draws sampler air through each tube. The flow rate is selected by setting an internal switch to draw either 10, 20, 30, 40, or 50 mL/min of air; the switch controls the on-off cycling rate of the pump over a 1-min period. A constant flow rate through each sample tube in the lid is regulated by a pressure sensing circuit located at the outlet side of the pump. The circuit is an integrator that supplies a voltage ramp to the pump motor, rising or falling as indicated by the outlet pressure. A flashing light-emitting-diode (LED), mounted on the BATS base control panel, gives a

visual indication that the pumping system is operating properly. This pumping system has proved to be more reliable than the originally installed pump, but consumes more power. Programmable controls are also placed on the base control panel which are used to control the number of samples, the sample duration, and to control either single or multiple sample start and stop times of a 7-day period. Two liquid crystal displays (LCDs), also mounted on the control panel, show the clock time, day of the week, and current tube number. A digital printer and integrated circuit memory module (Lagomarsino, 1989) record the start time, the day of the week, and the tube number for each sample. The BATS base controls are also used to assist in automated analyses when the lid is coupled to a gas chromatograph (GC).

For analysis, the perfluorocarbon tracers, retained on the Ambersorb adsorbent in the BATS tubes from the sampled air, are desorbed by resistance heating of the stainless steel tubes to 460°C. Current from the BNL gas chromatograph system (16.3 Amps AC) is supplied from a low voltage transformer (~1.55 VAC at the lid jacks) through the Canivalve solenoid assembly. The assembly consists of a 24-position rotary solenoid having two power decks capable of handling 20 amps. Twenty-three leads are wired to the power deck, each connected to the adsorbent tube floating clamp at one end of the respective tubes. The clamp at one end must float to allow for thermal expansion of the tube on heating (~0.8mm). A set screw secures the collar on the tube within the clamp; a similar set screw on a common aluminum rail secures the other end. Polyurethane rubber tubing (1/8-inch OD by 1/16-inch ID) is expanded over the 1/8-inch OD stainless steel adsorbent tubes and wire clamped to secure; the other end is attached to the Scanivalve 1/16-inch protrusions.

Tracer sample analysis

Tracer sample analysis will be done with a gas chromatography system. The gas chromatograph system is composed of a gas chromatograph, of data handling devices, gas standards, and a BATS. The Varian 6000 gas chromatograph consists of a series of specially designed traps, catalysts, columns, and an ECD-electron capture detector. The data handling system consists of an analogue electronic filter on the ECD electrometer output connected to a Nelson Analytical 300 Chromatograph system comprised of a Model 7653 Intelligent Interface and an IBM PC/AT with an ink jet printer and Nelson 2600 Chromatography Software. Brookhaven has also written extended software for further data processing and GC calibrations.

Analysis of a sample occurs when the sample is automatically thermally desorbed from the BATS sample tube. The sample is passed through a precut column and a Pd catalyst bed before being reconcentrated in an *in-situ* Florisil trap. Once the trap is thermally desorbed, the sample again passes through the same catalyst bed, another Pd catalyst bed, and then through a permeation dryer.

The sample is then passed into the main column where it is separated into the various perfluorocarbon constituents and then ultimately into the ECD for detection. Further details on the analytical system is given in Dietz (1987).

Release rates and expected crosswind average peak centerline concentrations at the Long Mesa and Hopi Point receptor locations are shown in the table below.

Summary of Expected Tracer Concentration					
Tracer Release			Receptor Site	Expected PFT levels	
Site	PFT	Rate, kg/h		PFT	Conc.[d], fL*/L
MPP	oPDCH	0.14[a]	Long Mesa	ocPDCH	4 ± 2
San Joaquin Valley	PTCH	0.14[b]	Hopi Point	PTCH	2 ± 1
Los Angeles Basin	PMCP	0.50[c]	Hopi Point	PMCP	14 ± 5

* fL = femto Liter = 10^{-15}L

[a] 100 kg for 30 days in January 1992 and 170 kg for 50 days in July-August 1992

[b] 70 kg for 21 days in July 1992

[c] 250 kg for 21 days in July 1992

[d] Crosswind average peak centerline concentration

At receptor sites, 12-hour tracer samples will be collected, and will sample 36 liters (L) of air. All other sites will sample 72 L over a 24-hour period. The table on the next page shows relevant information regarding the amounts of tracer expected, backgrounds, levels of detection, and signal to background ratios for the GC analysis. It can be seen that the limit of detection (LOD) and uncertainty are very small compared to background, except for PTCH, which has a limit of detection of about 16% of background and uncertainty of 50% of background. Thus, for the MPP and Los Angeles Basin tracers (ocPDCH and PMCP), even an additional tracer concentration of a fraction of background can be reliably quantified. For all 3 tracers, the uncertainty of the amount of tracer above background (signal-background) is small for expected crosswind plume centerline concentrations.

A sample chromatogram for a 20 L of ambient (background) air is shown in Figure 8. Background levels of the tracers used (ocPDCH, PMCP, and PTCH) can be clearly distinguished and quantified. PMCH, which is not being released, can be used as a reference.

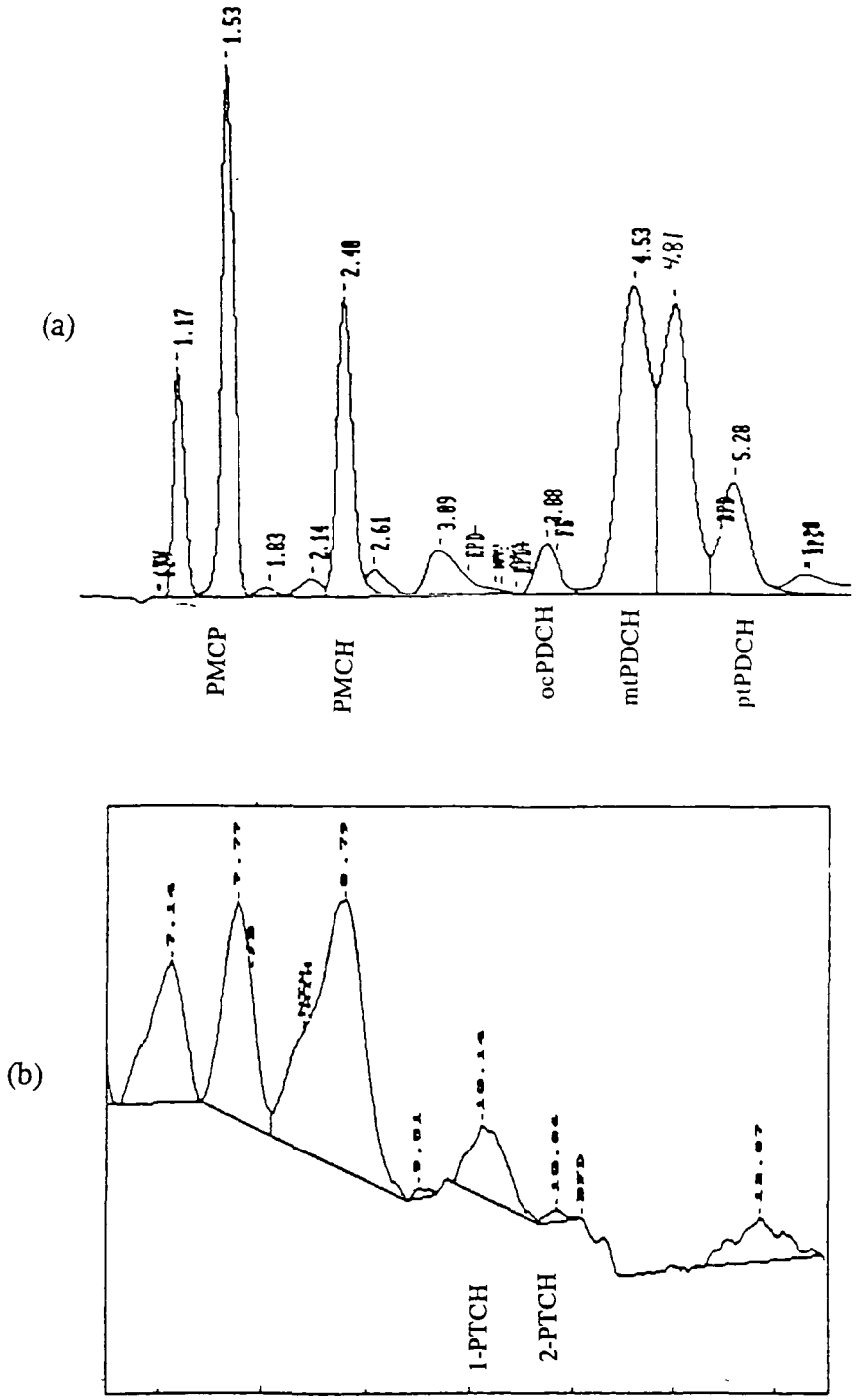

Figure 8. Chromatogram of 20 L sample of ambient air. (a) Elution time up to 6.5 minutes. (b) Elution time 6.5 to 13 minutes.

For 12 hour (36L) samples			
Background	ocPDCH = 0.3 fL*/L	PMCP = 3.3 fL/L	PTCH ≈ 0.1 fL/L
Area of Response (counts/fL)	360	160	300
Receptor concentration (fL/L)	4	14	2
Quantity in 36L (sample and background) fL	155	623	72
Quantity in 36 L background, fL	10.8	119	3.6
Limit of Detection, fL	0.05	0.12	0.6
Limit of Detection, counts	20	20	180
Uncertainty = 3 Limits of Detection, counts	60	60	500
Counts in 36L (sample and background)	55,800 ± 60	99,680 ± 60	21,600 ± 500
Counts in 36L background	3,888 ± 60	19,040 ± 60	1080 ± 500
Signal to background	14.35 ± 0.22 (± 1.5%)	5.24 ±0.017 (± 0.3%)	20.00 ± 9.27 (± 46.4%
Signal - background, counts	51,912 ± 85 (± 0.16%)	80,640 ± 85 (± 0.11%)	20,520 ± 707 (± 3.4%)

* fL = femto Liter = 10^{-15}L

Tracer Quality Control

Rates of air flow through the sampler are checked before and after the sampler is sent to the field monitoring site. This is to determine the total quantity of air sampled each sampling period. Adjustments are made to compensate for altitude and temperature differences. Three additional PFTs that are not released are used as a cross-check of the sampling volume. The concentration of these PFTs is essentially constant, so the quantity of air sampled can also be calculated from the amount of these tracers collected.

The sample analysis is done at 460°C. This is 50°C above the temperature needed to desorb all PFTs. After analysis, the sample tubes are "baked out" at 510°C to remove any remaining traces of PFTs. Before sending the tubes out to the monitoring sites, every fourth tube is analyzed. At this time, the tubes should have zero tracer. They are analyzed down to 30-50 counts,

which is about 1% of background. If a tube has zero signal, then it has not been sampled, because the ambient background has not been detected. The samplers will be programmed to collect 20 or 21 samples; tubes 22 and 23 should be zero.

5. Air Quality Monitoring

Purpose

Air quality monitoring for Project MOHAVE has many applications. The extinction budget analysis requires data for all the major particle components (e.g. sulfate, organic and elemental carbon, crustal, and liquid water as estimated by relative humidity) by particle size to be used in conjunction with optical data (scattering and extinction coefficients). The hybrid and receptor models need particle and gaseous sulfur concentrations and particulate trace elements as endemic tracers (such as arsenic for smelters) in addition to measurements of artificial tracer. Oxidants, especially hydrogen peroxide, should be monitored to assess the potential oxidant limitations of SO_2 to sulfate conversion. The air quality monitoring network will document the regional distribution of particulate and SO_2 and establish boundary conditions for the study area; used along with wind field information, transport of pollutants into the area will be identified. Eigenvector analysis of the pollutant fields will identify common patterns and may identify specific sources with the patterns. These data will also provide for a check of the deterministic modeling results.

IMPROVE Samplers

The IMPROVE sampler consists of four independent filter modules and a common controller, as shown in Figure 9. Each module has its own inlet, PM-2.5 or PM-10 sizing device, flow rate measurement system, flow controller, and pump. In the three PM-2.5 modules, the airstream passes through a cyclone that removes particles larger than 2.5 μm in diameter. The airstream then passes through a filter, which collects all the fine particles. In the PM-10 module, the inlet prevents particles larger than 10 μm from being sampled.

Channel A collects fine particles (<2.5 μm) on a Teflon filter and provides total fine mass, elemental analysis (H and Na-Pb), and absorption. Particle Induced X-Ray Emission (PIXE) analysis gives the concentration of the elements Na-Pb; Hydrogen is obtained by Proton Elastic Scattering Analysis (PESA). Absorption is determined by the Laser Integrating Plate Method (LIPM).

Channel B uses a fine nylon filter behind a nitrate denuder for ion chromatography analysis (Cl^-, NO_2^-, NO_3^- and SO_4^{2-}). Channel C is used to obtain organic and elemental carbon from a fine quartz filter. A thermal/optical carbon analyzer which makes use of the preferential oxidation of organic and elemental carbon compounds at different temperatures is used. Channel D measures PM-10 total mass on a Teflon filter and SO_2 with an impregnated quartz filter. More detailed descriptions of the IMPROVE samplers, analysis methodologies, and protocol appear in Pitchford and Joseph (1990), and Eldred

et al. (1988). The location of sites and monitoring schedules for IMPROVE samplers is shown in Section 3.

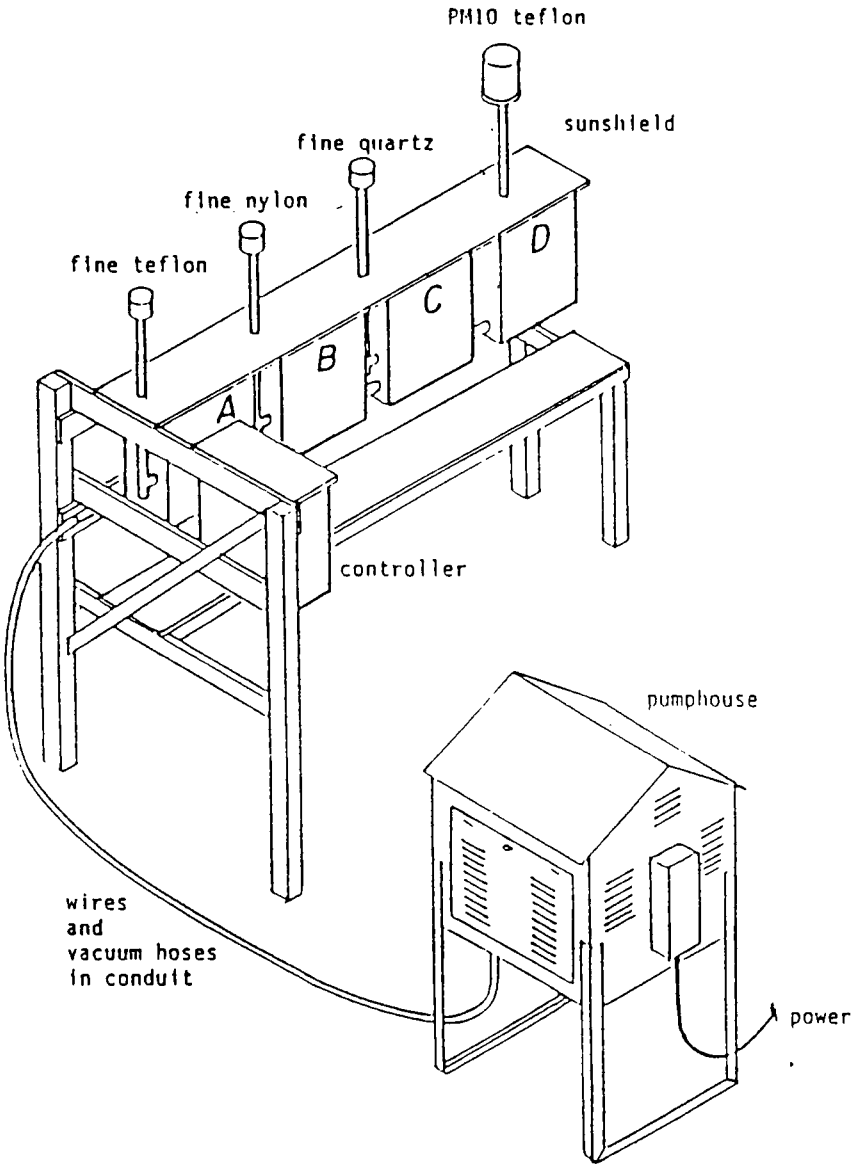

Figure 9. Schematic of IMPROVE sampler.

DRUM Samplers

DRUM (Davis Rotating-drum Universal-size-cut Monitoring) samplers will be used at six locations. The DRUM particulate samplers partition the aerosol into eight size ranges. This provides critical information to relate aerosol to extinction because of the strong relationship between particle size and light scattering. PIXE and PESA analysis is done to determine the concentration of

each element (H and Na-Pb) by size range. The size distribution, hence the light scattering efficiencies, for different particulate component species can be inferred, if sufficient material is collected (e.g. see Cahill *et al.*, 1987). The DRUM sampler is described by Raabe *et al.* (1988).

Six DRUM samplers will be deployed. The sampling time will be either four or six hours. The receptor sites will have DRUM sampling for the entire study; the remaining samplers will be placed at other locations of interest yet to be identified. Among the possible sites are Tehachapi Pass, Cajon Pass and Spirit Mountain. Analysis of all samples is beyond the resources of Project MOHAVE. All samples will be archived; analysis of samples will be done for selected periods of interest.

High Volume Dichotomous Samplers

High volume (300 L/minute) dichotomous samplers will be used to improve the trace metal data base for receptor modeling with endemic tracers. Aerosols in the size ranges 0.05-2.5 μm and 2.5-20 μm will be collected on Teflon filters. Instrumental Neutron Activation Analysis (INAA) and X-Ray Florescence (XRF) will be done on the samples. Three samplers will be used. One sampler will be equipped with a trap to collect semi-volatile organics. The locations have not been decided yet; one will characterize background and one will be near the mouth of the Grand Canyon.

Scanning Electron Microscopy (SEM) will be used to characterize individual particle morphology and elemental composition. In addition, Computer-Controlled SEM (CCSEM) analysis can be used to increase the numbers of particles analyzed and eliminate possible human operator microscopy bias. CCSEM data can then be used in data analysis approaches that require quantitative composition as a function of particle size and shape distributions. Mie Theory calculations to determine extinction budgets can use this data. Unlike other data sets, CCSEM data allow for direct analysis of the questions of aerosol mixture (i.e., the extent to which component species are constant in all particles in an individual sample). Receptor models can be based upon an endemic tracer approach using CCSEM data, or it can use the individual particle characterization information to aide in resolving issues raised by other attribution approaches.

Hydrogen Peroxide Measurements

Hydrogen peroxide (H_2O_2) is likely to have a significant role in the formation of sulfate in the study region when clouds are present. Aqueous phase conversion of SO_2 to SO_4^{2-} is critically dependent upon hydrogen peroxide (H_2O_2) and ozone (O_3) (Penkett *et al.*, 1979; Calvert *et al.*, 1985). Hydrogen peroxide is thought to be the leading oxidant of dissolved SO_2 in the eastern United States, where the pH of atmospheric water is generally below 4.5 (Heikes *et al.*, 1987). In the desert southwest, where the pH of atmospheric water is typically higher,

ozone may also be important in the aqueous phase oxidation of SO_2. Saxena and Seigneur (1986) also identify O_2 catalyzed by Fe^{3+} and Mn^{2+} as an important aqueous phase oxidant of SO_2. Hydrogen peroxide reaction rates with dissolved SO_2 are typically 50-100% per hour (Lee *et al.*, 1986); thus the presence of clouds with sufficient H_2O_2 present can result in rapid sulfate formation. The amount of hydrogen peroxide available for oxidizing SO_2 may be limited, especially during winter, when photochemical generation of H_2O_2 is low (Calvert *et al.*, 1985; Kleinman, 1986).

The NAS review of WHITEX noted that H_2O_2 measurements were not made; the NAS used values measured in Tennessee (about the same latitude as GCNP) to estimate potential sulfate formation. Members of the Committee on Haze in National Parks and Wilderness Areas suggested that Project MOHAVE make some measurements of hydrogen peroxide. If measurements of hydrogen peroxide show sufficient amounts to convert all the SO_2 to sulfate, we can likely conclude the atmosphere is not oxidant limited. However, showing that molar quantities of hydrogen peroxide less than sulfur dioxide does not necessarily indicate oxidant limiting conditions. Ozone effects may be significant if the pH is adequately high. Heikes *et al.*, found that SO_2 concentrations were a factor of 3-5 greater than H_2O_2 concentrations in the surface layer, but above the surface layer H_2O_2 concentrations were twice the SO_2 concentrations. Even with aircraft vertical profile measurements, Heikes *et al.* concluded that the hydrogen peroxide measurements were ambiguous in determining if the atmosphere was oxidant limited. Their near cloud observations suggested that physical-dynamical processes may be as or more important than a simple molar comparison of SO_2 to H_2O_2 at ground or cloud level.

It is not possible for Project MOHAVE to fully characterize the temporal and spatial distribution of atmospheric hydrogen peroxide necessary to conclusively determine oxidant limitations. However, limited measurements may provide some insight into the potential for hydrogen peroxide oxidation of SO_2. As in the NAS report, sulfate concentrations may be compared to H_2O_2 concentrations to see if sufficient H_2O_2 existed to account for the measured sulfate values. Project MOHAVE will make a limited number of hydrogen peroxide measurements. The SRP NGS study made hydrogen peroxide measurements during the winter of 1990. These measurements may be used to estimate H_2O_2 for the winter intensive.

Methylchloroform Measurements

Methylchloroform has been identified as a tracer of weekday emissions from the Los Angeles Basin (White *et al.*, 1990). Miller *et al.* (1990) found that methylchloroform levels at Spirit Mountain are correlated with particulate light scattering, with the majority of hazy conditions having elevated methylchloroform levels. Methylchloroform measurements, in conjunction with meteorological data and modeling, can aid in identifying periods when air previously in the Los

Angeles Basin is in the study area. However, a limitation of methylchloroform as a Los Angeles Basin tracer is that the emissions are primarily weekday emissions, with weekend emissions being much lower. Thus weekend emissions from the Los Angeles Basin might not be tracked using this tracer and the absence of methylchloroform does not necessarily indicate an absence of air from the Los Angeles Basin.

Desert Research Institute will measure methylchloroform at Spirit Mountain, Meadview, and Long Mesa. These data will be investigated for use in identifying the presence of air previously in the Los Angeles Basin. During the summer intensive the release of perfluorocarbon tracers from the Los Angeles Basin and San Joaquin Valley should provide a check on the utility of methylchloroform as a Los Angeles Basin tracer.

6. Meteorological Monitoring

Background

Meteorological monitoring is necessary to characterize the speed, direction, and depth of transport in the region and for model initiation and validation. The existing National Weather Service (NWS) surface and upper air monitoring sites are insufficient to characterize the complex meteorological setting of the study area. In addition, NWS upper air measurements (rawinsondes) are taken only twice per day. Thus, they may not capture important small time scale meteorological changes and because they provide nearly instantaneous measurements, they may not be representative of average conditions.

The Wave Propagation Laboratory (WPL) of the National Atmospheric and Oceanic Administration (NOAA) will provide much of the meteorological measurements data for Project MOHAVE. Air Resource Specialists (ARS), the optical monitoring contractor, will provide surface meteorological data at the four receptor sites. WPL has a unique capability of providing continuous wind and temperature profiles in the atmospheric boundary layer (ABL) using wind profiling radars with Radio Acoustic Sounding Systems (RASS). The radars transmit 915 MHz signals and receive back-scattered signals from the atmosphere. With three antennas, usually two tilted and one vertical, the three components of the wind can be measured using the Doppler effect. The best results are obtained when the winds are averaged over about one hour. The RASS component uses the Bragg scatter of radar waves from vertical propagating acoustic waves to measure the sound speed. Because the sound speed depends upon air temperature, temperature profiles can be derived. Usually the instrument is configured to provide one 5-minute averaged temperature profile each hour. The backscattered intensities received by the wind profiler in the form of signal-to-noise ratios can also qualitatively indicate mixing depths. The advantage of the wind profiler/RASS instruments over rawinsondes is that they provide continuous profiles in time.

Objectives

The wind profiler/RASS data consist of wind profiles, nominally to 2.5 km, and temperature profiles to almost 600 m. These data are necessary to characterize the speed, direction, and depth of material transport in the region and also necessary for model initiation and validation. The primary objective is to measure the transport of material from the MPP to GCNP. Also, it is important to characterize the flow from major urban areas in the region (e.g., South Coast Basin, Las Vegas, Phoenix/Tucson) and to separate this flow from flow containing the MPP emissions. There are two other major power plants nearby, the Reid Gardner Plant near Overton, Nevada, to the northwest of the Grand Canyon and the NGS near Page, Arizona to the northeast. It is also desirable to

determine the frequency of transport from these sources.

There are several ancillary problems which relate to the potential transport paths which the MPP plume may take into the Grand Canyon region. An indirect path is along the Colorado River to the north and then over Lake Mead. Because the lake is lower in elevation than the surrounding terrain and the ABL over the lake is usually more stable than that over the surrounding land due to the relatively cool water, the pollution may pool and collect in the Lake Mead Basin. Also, material from other sources (e.g. Las Vegas/Henderson or Reid Gardner) may collect in the same basin. A change in wind may transport this material into the lower portion of the Grand Canyon near Meadview. It is therefore important to monitor the winds and stability in the Lake Mead area, both over the lake and near the western entrance to the Grand Canyon.

With southwesterly flow near the surface, the material from the MPP may be transported more directly toward the Grand Canyon region over the high plateaus to the northeast of the plant. This path also requires meteorological monitoring.

Surface meteorology will be monitored at all the wind profiler sites so that the lower gates of the profilers can be compared with surface parameters. Also, NOAA/WPL will provide at least four surface pressure sites. Gaynor *et al.* (1991) have shown that winds calculated from surface pressure gradients can be used as surrogates for transport winds. The pressure array will allow calculations of mesoscale transport winds which can be compared with and be adjunct to profiler winds.

Another contribution from NOAA/WPL will be the wind and temperature data from profiler/RASS operations performed as part of the South Coast Air Basin study beginning in July, 1992, and continuing through the summer intensive period. Among the tentative locations for these instruments are the Cajon, Banning and Tehachapi passes. These data will be useful adjuncts to Project MOHAVE by providing upstream information on potential transport from the Los Angeles Basin and San Joaquin Valley into the MPP region. Starting in February, 1992 wind profiler data from sites on the Mogollon Rim (central Arizona) will be available. These wind profiler sites will help characterize periods of flow from the southeast into the study area.

NOAA/WPL will also provide tethersonde and airsonde profiles for short periods and at critical locations during the winter and summer intensives. These profiles will be measured at transport, drainage, or pooling locations that will not have regular continuous measurements. Because of the limited height range of the tethersonde, the preferred locations for these profiles will be in regions with shallow boundary layers. One general area of this type is the Lake Mead basin where a relatively shallow ABL compared to the surrounding desert may persist well into the morning due to the cool water surface and to the nocturnal drainage of cool air into the basin.

Field Study Plan

A wind profiler with RASS will operate in close proximity to the MPP from September 1991 through September 1992. This location will be supplemented with the DRI operation of an AeroVironment Doppler sodar for a quality control (QC) check of the profiler and to supplement the profiler with detailed low level winds. Another wind profiler with RASS will be located at Truxton to monitor the possibility of direct southwest to northeast transport from the plant. The Truxton site is in open terrain; this allows the data from this site to reflect the general flow patterns over the entire study area. It will also measure the south and southeast summer monsoonal flow from which directions material may be transported from Phoenix and Tucson or from smelters to the south and southeast. A doppler sodar will also operate most of the study period at Meadview.

In support of the winter intensive study, two additional wind profilers will be operated from mid-November 1991 through late-January 1992. The site locations will be the following:

1) South of MPP in the vicinity of Needles, which is usually downwind of MPP during the winter.

2) At Temple Bar, on the south shore of Lake Mead, about 30 km west of GCNP. This site will help characterize low level flow over Lake Mead, which may vary significantly from the flow at higher levels.

During the winter intensive period, NOAA/WPL will intermittently operate a tethersonde and/or radiosonde to supplement the upper air data. The locations may be at Cottonwood Cove to monitor wind and stability in the upper Mohave Valley, or near Lake Mead to monitor the meteorology in the Lake Mead Basin.

From July 1992 through September 1992, a supplemental profiler will operate at Cottonwood Cove (Lake Mohave) in support of the summer intensive experiment. The plume is typically transported past this site, especially during night and morning hours, and may exit the Colorado River valley near this site during the late morning and afternoon. An additional wind profiler will be located at Meadview. The sodar at Meadview will be moved to Temple Bar to measure low level flow above Lake Mead. The combination of doppler sodar at lake level, combined with a wind profiler at Meadview, 500 meters above lake level, will provide a vertical profile extending to about 3 km above lake level. NOAA/WPL will likely participate in the South Coast Air Basin Study which will occur during the same period as the MOHAVE summer intensive. WPL will have six profilers operating in the South Coast Basin. One or two of those will be on the east (desert) side of the Tehachapi, Cajon, or Banning Passes. Combining data from the South Coast profilers with data from the profilers

deployed for the MOHAVE summer intensive will provide a rare opportunity to continuously monitor the winds from Southern California to the Grand Canyon.

Data Collection

All the profilers will provide hourly consensus averaged winds in two modes -- a high range resolution mode, usually about 60 to 100 m and a low range resolution mode, usually 200 to 400 m. Minimum heights of around 150 m and maximum ranges of about 2.5 km are expected. During the more moist summer monsoon period, much higher ranges may be expected.

The RASS temperature profiles are measured once per hour representing 5 minute consensus averaged profiles. The minimum range is about 150 m; the maximum range expected under dry desert conditions is nominally 600 m. The Doppler sodar at Overton or Temple Bar will provide a minimum range of about 50 m and a maximum range of about 600 m with about a 50 m range resolution.

The surface meteorological data associated with the profilers will probably represent 5 minute averages of wind, temperature, and relative humidity measured about 3 m above the ground. The locations measuring surface pressure will also have temperature and relative humidity instrumentation.

Where phone lines are available, all profilers, including those with RASS, and the sodar will be interrogated by phone once per day and the ASCII files sent to a hub work station located at NOAA/WPL in Boulder, Colorado. This validation level zero data will also be available at each site from printer paper and on the hard disks of each controlling PC. The surface meteorological data collected at the profiler sites will also be sent over the same phone lines to the hub. The pressure sites, unless co-located with the profilers, may not have phone line capabilities depending on the feasibility of installing lines.

Data Quality Assurance

Wind profilers and Doppler sodars identical to those to be deployed for Project MOHAVE are periodically tested and compared at NOAA's Boulder Atmospheric Observatory which includes a 300 m meteorological tower. The RASS derived temperatures are also compared to thermometers on the tower. All instrumentation will have been previously tested in other field studies prior to deployment. The collocation of a Doppler sodar at the MPP with a wind profiler will provide a continuous field quality assurance check on both the profiler and the sodar.

All the data that is recorded and printed out at each site and sent over phone line to the hub in Boulder will be level zero. The field programs on each control computer for the radar/RASS and sodar provide consensus averaging which is equivalent to on-line, real-time sorting of data according to consistency criteria. The wind profiler/RASS and sodar data will be screened by an automated editor (Wuertz and Weber, 1989) after each 24 hour collection period.

This data will in turn be inspected by qualified staff and flagged if required. The resulting ASCII files of winds and temperatures, along with graphical displays, will then be available for quick dissemination by diskette or by electronic transfer.

The in situ surface meteorological data will require similar inspection and will be averaged into one hour blocks. These data will be available for similar dissemination.

Data Processing and Analysis

The senior scientific staff at NOAA/WPL will cooperate closely with the modelers to ensure that level one data are readily available to them in a useful form. NOAA/WPL scientific staff will take leadership in analyzing wind profiler/RASS, sodar, tethersonde, radiosonde, and surface meteorological data to gain insight into the often complex transport processes in the project region. This effort will require the use of various types of data from project collaborators outside of NOAA. The surface pressure array may be very critical in extending the understanding of material transport over a larger area than that covered by upper air wind measurements.

7. Optical Monitoring

Overview

The optical monitoring plan for project MOHAVE consists of two fundamental aspects:

1) View Monitoring

View monitoring documents the visual impairment of specific unique vistas under various air quality conditions. View monitoring is primarily accomplished with 35mm color slide photography and 8mm color time-lapse photography. Color slides provide high resolution documentation of the visible effects of uniform and layered hazes on the vista. Digitization of the slides can be done to yield relative radiance fields that can be used to calculate color contrast, average landscape contrast, visual range, modulation depth, equivalent contrast, and just noticeable change. In addition, slides of extremely clean days can be used as the basic input to present visual air quality scenarios. 8mm time-lapse photography captures the important spatial and temporal patterns of visibility events that allow for a more in-depth understanding of visual air quality.

2) Electro-Optical Monitoring

Electro-optical monitoring measures the basic electro-optical properties of the ambient atmosphere and aerosols, independent of specific vista characteristics. Monitoring will include measurements of the ambient atmospheric extinction coefficient (b_{ext}), and its scattering (b_{scat}) and absorption (b_{abs}) components. Primary operational monitoring techniques include the transmissometer (b_{ext}), nephelometer (b_{scat}), and filter absorption (b_{abs}). Temperature and relative humidity measurements, taken simultaneously with electro-optical measurements, are mandatory to infer visibility effects associated with chemical and physical interactions between water vapor, liquid water, and aerosols.

Project MOHAVE will incorporate current state-of-the-art monitoring instrumentation, operating and quality assurance procedures, and data collection, reduction, editing, and reporting protocols that have been developed for the IMPROVE monitoring program (ARS, 1990a; ARS 1990b).

View Monitoring

Equipment

Automatic 35mm and 8mm camera systems will be an integral part of the optical monitoring for project MOHAVE. The spatial and temporal variations in visual air quality captured by these systems will be used to:

- Document how vistas appear under varied conditions;
- Qualitatively record the frequency that various conditions occur; e.g. incidence of uniform haze, layered haze, plumes, and meteorology;
- Provide a quality assurance reference for collocated electro-optical measurements;
- Serve as a backup method to estimate the electro-optical properties of the atmosphere (if appropriate teleradiometric targets are in view);
- Support the calculation of advanced visibility indices;
- Support computer imaging studies;
- Provide quality media for visually presenting program goals, objectives, and results to study participants, decision makers, and the public.

Systems based on the following cameras will be used:

- 35 mm cameras: Olympus OM series
  Contax 136 and 167
  Cannon EOS series

- 8mm time-lapse: Minolta 601 series

Standard operating procedures developed for the IMPROVE monitoring program will be followed (ARS, 1990a).

Monitoring Locations and Sampling Frequency

The 35mm camera systems will be located at all receptor sites and other Class I Area sites. The 8mm time-lapse systems will be located at Meadview and various scenic view points along the south rim of the Grand Canyon. During non-intensive periods, only 35mm cameras will operate, taking three exposures daily at 0900, 1200, and 1500 hrs.

During intensive monitoring periods, 35mm cameras at the receptor sites and at GCNP will take nine exposures daily from 0800-1600 hrs. Time-lapse photography will take 1 frame per minute from 0800-1600 hrs daily. Additional

view monitoring locations will be added as the study progresses.

Electro-optical Monitoring

Extinction Measurements

The Optec, Inc. LPV-2 long path transmissometer will be the primary instrument used to measure b_{ext} for project MOHAVE. The transmissometer incorporates a light detector (receiver) at one end of a specific atmospheric sight path. The receiver directly measures the illuminance of a constant output light source (transmitter) located at the opposite end of the path. Calibration of the transmissometer accurately determines the inherent output of the transmitter. The transmission of the sight path can then be calculated:

$$T = \frac{I_r}{I_{cal}}$$

where

T = transmission of sight path r
I_r = illuminance measured by receiver at distance r
I_{cal} = calibration illuminance of transmitter

By measuring the exact length of the sight path the average atmospheric extinction coefficient of the path can be calculated:

$$B_{ext} = \frac{-\ln(T)}{r}$$

where

b_{ext} = average extinction coefficient of sight path r
T = transmission of sight path r
r = length of sight path r

During the past ten years, transmissometers have been developed, tested, and deployed in the IMPROVE monitoring network, National Park Service IMPROVE protocol sites, and various other monitoring programs. They have become the accepted method for reliably making continuous precise, accurate, b_{ext} measurements. Standard operating and data reporting procedures developed for the IMPROVE program will be followed (ARS, 1990b).

Scattering Measurements

Integrating nephelometers will be used to measure b_{scat}. The integrating nephelometer measures b_{scat} by directly measuring the light scattered by aerosols and gases in an enclosed sample volume. The scattered radiation is integrated over a large range of scattering angles. Since the total light scattered out of a sight path is the same as the reduction of light along the sight path due to scattering, a properly calibrated integrating nephelometer gives a direct measurement of b_{scat}.

Nephelometer measurements are involved in considerable controversy because of the modification of the ambient aerosol as it passes through the sampling train and optical chamber. The instrument heats the air thus lowering the relative humidity environment of the aerosols. This leads to an underestimation of ambient b_{scat}. Extreme efforts have been made to operate nephelometers as close to ambient temperatures as possible. The best results have been a heating of approximately 1.5° C. This is approximately a 10% change in relative humidity, which can lead to underestimation of ambient b_{scat} measurement. In addition, nephelometers underestimate the scattering by coarse particles ($> 2.5\mu m$ in diameter). As with the transmissometers, standard protocols developed for the IMPROVE program will be followed (ARS, 1990b).

Absorption Measurements

Where collocated transmissometers and nephelometers are collecting data, b_{abs} will be estimated by subtracting b_{scat} from b_{ext}. The term, b_{abs}, will also be estimated by absorption measurements from channel A filters collected by the aerosol monitoring network. These b_{abs} measurements will be average values for the collection period of each filter. Data from these measurements will be available only for periods when aerosols measurements are taken.

Temperature and Relative Humidity Measurements

Accurate air temperature and relative humidity data are critical to establish the relationship between ambient aerosols and visibility effects. Small changes in relative humidity, especially above 70%, can dramatically affect aerosol size and optical characteristics. Rotronic Instrument Corporation Model MP-100F sensors will be used in Project MOHAVE. The MP-100F combines a 100 ohm platinum temperature sensor with an enhanced hygroscopic polymer film humidity sensor to provide an integrated air temperature/relative humidity device that will maintain a 2% relative humidity measurement accuracy over the range of 0-100% relative humidity. These sensors will be operated with every transmissometer and nephelometer in the Project MOHAVE network.

Monitoring Locations and Sampling Frequency

Transmissometers and nephelometers will operate continuously through the year of Project MOHAVE at various locations. Data from instruments specifically installed for Project MOHAVE as well as data from other existing networks will be collected for inclusion in the Project MOHAVE data base. Measurements from the following sites, with sponsoring networks, are listed in the table below. Data will be collected and archived as hourly averaged values for the entire monitoring year.

Transmissometer and Nephelometer Monitoring Locations in the Southwest			
Site	Sponsoring Network	Transmissometer	Nephelometer
Bandelier NM	NPS	✓	
Big Bend NP	IMPROVE	✓	
Bryce Canyon NP	SRP		✓
Canyonlands NP	IMPROVE	✓	
Chirichahua NM	NPS	✓	
Grand Canyon NP			
south rim	IMPROVE/SRP	✓	✓
in-canyon	NPS	✓	
Long Mesa	SCE		✓
Meadview	MOHAVE/SCE	✓	✓
Mesa Verde NP	IMPROVE	✓	
Page, Arizona	SRP		✓
Petrified Forest NP	NPS	✓	
San Gorgonio W	IMPROVE	✓	
Spirit Mt., Nevada	SCE	✓	✓
Tonto NM	IMPROVE	✓	
Guadalupe Mts. NP	NPS	✓	
Total		13	6

8. Emission Inventory and Characterization

Purpose

Emission inventory and source characterization are necessary for the deterministic and receptor modeling. Receptor models need source characterization for the main sources of interest. This involves compiling a ratio of elements that uniquely identifies a source and can be monitored at the receptor sites. The emission inventory is used to supply input to the deterministic modeling. Emission inventory consists of quantifying the emission rates of substances of interest from all sources that may be reasonably expected to impact the study area. For Project MOHAVE, sulfur dioxide emissions are of the greatest interest. The SO_2 emissions from MPP will be modeled with the transport and chemical models described in section 10. The level of modeling of other sources is still being investigated. Project MOHAVE intends to include transport and first-order chemical modeling of other significant sources of SO_2, including the southern San Joaquin Valley, the Los Angeles Basin, other powerplants, and copper smelters within the domain of the meteorological modeling area. The source profiling will also detail the primary particle emissions in order to assess whether primary particles contribute significantly to extinction.

Review of Existing Data and Inventories

The emission inventory used in the SRP NGS study (Systems Applications International, 1991) will be reviewed. State air pollution agencies will be consulted about emission data, especially regarding any changes for the main sources of SO_2. The power output of the MPP will be used to determine the emissions from the MPP. The operational status of other large SO_2 sources will also be checked and emission rates adjusted if necessary before the modeling analysis.

MPP Stack Sampling

Stack sampling will be done to determine the composition and quantity of MPP emissions, which are needed for the receptor, hybrid and deterministic modeling analyses. This component of the study has not yet been planned. The study plan will be updated when details of the stack sampling are known.

9. Centralized Data Management and Validation

Overview

EPA/EMSL in Las Vegas will be the data managers for Project MOHAVE. Information will be obtained from the following sources:

SOURCE	DATA TYPE
NOAA/WPL	Surface and Upper Air Meteorology
Brookhaven National Laboratory	Tracer concentrations
UC-Davis	Aerosol and SO_2
Air Resource Specialists	Optical and Surface Meteorology
EPA-RTP/AREAL	Aerosol
National Meteorological Center	Surface and Upper Air Meteorology
Colorado State University	Meteorological Modeling
CAPITA- Washington University	Monte Carlo Modeling

The data to be collected are described in more detail in Sections 3-7. A data management and validation plan will be developed by the data management coordination committee. A sketch of the expected elements of the plan to be developed is presented in the remainder of this section.

Two levels of validation (Levels 1 and 2) will be systematically applied. Level 1 (univariate) validation involves checking the data for outliers, rates of change, proper indication of time and location of data, etc. In Level 2 (multivariate) validation, consistencies among variables and the appropriateness of spatial and temporal patterns are investigated. For example, the light scattering (b_{scat}) measured by a nephelometer should be less than the total extinction (b_{ext}) obtained by a transmissometer. Level 3 validation occurs during the data analysis. If data inconsistencies are found, the documentation regarding the questionable observation is examined for correctable errors (e.g. transcription errors). Uncorrectable, suspect data are flagged, but not removed from the data set. Data known to be incorrect and not recoverable are removed from the data set.

Each group responsible for collecting data will perform at least Level 1 validation. UC-Davis will do a partial Level 2 validation of the aerosol data. The data managers at EPA/EMSL Las Vegas are responsible for the Level 2

validation. Systematic procedures and protocol for the Level 2 validation will be developed and fully documented prior to releasing the data. Level 1 protocols utilized by organizations responsible for each data subset will also be documented. A computerized listing of the data will be prepared. Level 2 data will be distributed to data analysts and other interested parties. At the end of the study, all data will be assembled and documented. A brief discussion of validation conducted by some of the participants appears in the following subsections.

Aerosol Sampling (UC-Davis)

A number of the measured or derived parameters are interrelated. This allows data intercomparisons as a method to evaluate system performance and check for outliers. The intercomparisons made are listed below:

(1) Fine sulfur vs. fine sulfate
(2) Fine sulfur vs. PM-10 sulfur
(3) Fine hydrogen vs. fine mass
(4) PM-10 hydrogen vs. PM-10 mass
(5) Sum of fine components vs. fine mass
(6) Sum of PM-10 components vs. PM-10 mass
(7) Elemental carbon vs. optical absorption
(8) Organic carbon vs. nonsulfate hydrogen
(9) Fine mass vs. extinction
(10) PM-10 mass vs. extinction
(11) Fine mass components vs. extinction
(12) PM-10 mass components vs. extinction

Details of the quality assurance and data validation are given in Pitchford and Joseph (1990).

Transmissometer Data

The transmissometer data is subjected to three levels of validation. In the first level, validity codes reflecting transmissometer instrument operation are added to the raw transmissometer data files. In the second level, data and validity codes are checked for inconsistencies using a screening program. The b_{ext} data are adjusted for lamp drift of 2% per 500 hours of lamp-on time. Validity codes are added to all data. The third level, consists of 2 steps,

(1) Calculation of uncertainty values for all data; and

(2) Identification of b_{ext} values affected by weather.

Validity codes for b_{ext} include:

0 = valid
1 = Invalid: Site operator error
2 = Invalid: System malfunctioned or removed
3 = Valid: Data reduced from alternate logger
4 = Weather: Relative Humidity > 90%
5 = b_{ext} > maximum threshold
6 = Δb_{ext} > delta threshold
7 = b_{ext} uncertainty > threshold
8 = Missing: Data acquisition error
9 = Invalid: b_{ext} below Rayleigh
A = Invalid: misalignment
L = Invalid: Defective lamp
S = Invalid: Suspect data
W = Invalid: Unclean optics

Radar wind profilers and RASS

Real-time processing consists of a Doppler spectra peak picking routine which searches for spectra peaks beginning from the highest level of good signals to the lowest gate. As the routine searches for peaks in a downward direction, it requires consistency from gate-to-gate. If a peak shifts beyond a given threshold between gates, that peak is rejected. To help eliminate ground clutter, the algorithm also rejects peaks near zero velocity if a secondary peak away from zero is available. After the peaks, or first moments, for each individual radial are chosen in this way, a consensus averaging is performed. This technique requires at least 50% of the points on each gate of each radial for a 55 minute period (5 minutes for RASS temperature) to fall within a bin of 2 m/s in width before the individual points in the bin are averaged. If less than 50% of the points fall within the bin, the radial component is flagged as bad and is not available for that period. A similar technique is used for the RASS derived temperatures with a bin threshold of 1 °C.

The normal post-processing quality assurance procedures consist of applying a time/height editor, normally referred to as the Weber/Wuertz editor (Weber and Wuertz, 1989), to each 24 hour period of one hour averaged profiler wind data or 5 minute averaged temperature data (one 5 minute average provided each hour). The editor assesses the neighborhood of each point for consistency in both speed and direction (or temperature), allowing for a larger tolerance for direction differences at lighter wind speeds. The tolerances are adjustable and depend on the prevailing meteorology during a particular experiment. The neighborhood size is also adjustable, but usually the eight adjacent points are chosen, if available. This editor has proven to be very powerful in eliminating outlier points. The results of this processing provide the Level 1 data.

NOAA/WPL is experimenting with applying a more sophisticated form of this editor on the radial moments before performing an hourly average. The test data are from the 1990 San Joaquin Valley Air Quality Study. The technique requires considerable processing. The decision to use this technique for Project MOHAVE depends on the quality of the data, which in turn depends on site characteristics.

The post-processed, Level 1 data will be compared with optically tracked rawinsonde (airsonde) wind and temperature profiles measured at each location. Several rawinsonde profiles will be available at each of the wind profiler locations representing different stability and meteorological conditions at each site.

10. Descriptive Data Analysis and Interpretation

Goals

A large quantity of data will be collected in support of Project MOHAVE. The descriptive data analysis and interpretation component of the study is intended to summarize the main features of the data as well as especially interesting cases, and offer physical explanations whenever possible. In contrast to the attribution analyses described in Section 11, this section will organize the data in a manner that will allow inference of effects from different sources, but will not generally be quantitative sufficiently to permit source apportionment.

Descriptive Statistics

Descriptive statistics will include calculation of means, standard deviations, skewness, and extreme values of the variables. In addition, time series of the data will be presented. These will include time series of the extinction coefficient (b_{ext}), tracer, sulfate, nitrate, organics, light-absorbing carbon, fine soil, and various trace elements and meteorological variables, for example. Correlations between variables will also be calculated.

Extinction Budget

Light extinction is caused by scattering and absorption by particles and gases. In general, particle scattering is the primary component of extinction, although in remote areas of the southwest, scattering by gases that compose the atmosphere (Rayleigh scattering) is a significant fraction on the clearest days. Black carbon (from diesel engines, forest fires, etc.) is the principal agent of particle absorption, and is occasionally an important contributor to haze in the study region. NO_2 is the only common gaseous pollutant that absorbs in the visible portion of the spectrum and is not likely to be a significant contributor to haze in GCNP.

The extinction budget analysis involves determining the contribution to extinction by all the major aerosol components. There are two fundamentally different approaches to estimate the extinction budget. A statistical approach uses multivariate analysis to explain the optical parameter (b_{ext} or b_{scat}) by a linear combination of the components. These components are the concentrations of the pollutant species (e.g., crustal, sulfate, nitrates, elemental and organic carbon, etc.) multiplied by best-fit determined coefficients interpreted as extinction efficiencies. The hygroscopic particle species (e.g., sulfate and nitrate) include a function of relative humidity to incorporate the effects of water upon the extinction efficiencies of these species.

An externally mixed aerosol (i.e. separate aerosol components are not contained within the same particles; for example, sulfate-coated crustal particles

would not constitute an external aerosol mixture) is implicitly assumed by the statistical approach for extinction budget analysis. The extent to which this assumption is true, and the implication of it being violated, are hard to estimate in any individual situation. In general, the greatest impact of non-external mixtures is thought to be associated with an interpretation of how changes in aerosol composition would affect atmospheric optics. In other words, there is increased uncertainty associated with the prediction of how visibility will respond to changes in emission caused by violation of this assumption.

In addition to the concern about implied assumptions, any use of multivariate statistics carries with it the concerns caused by use of possibly highly covariant independent parameters, and the use of measured parameters with large differences in relative measurement uncertainty. Both of these concerns can result in biased results. However, there are standard approaches to detect and minimize the impacts of these concerns.

The other approach to estimating extinction efficiencies for the various aerosol components is by first principle calculations (Mie Theory). These calculations require as input, certain particulate characteristics such as the distribution of particle size, shape, and indices of refraction. Generally, the size distribution can be estimated from size segregating sampler measurements. For Project MOHAVE, this will be done with the DRUM impactors for some components, such as sulfur and crustal components, but not for others, such as organic carbon and nitrate species. A functional relationship between water and the hygroscopic particles must be assumed to estimate its effects on particle size. Particle shape is generally assumed to be spherical, and the refractive indices are assumed to be the same as the bulk indices for the various measured particle chemical components. Assumptions must also be made concerning the nature of the aerosol mixture (i.e., external, internal, or some combination) in order to calculate the extinction efficiencies of the components. The extent to which these deficiencies and assumptions affect the calculated extinction is unknown.

In spite of the uncertainties discussed above, extinction budget analysis done by the two approaches generally results in similar extinction efficiencies. Since Project MOHAVE will use both Mie Theory and statistical approaches, the results can be intercompared for consistency, and reconciled with extinction efficiency values from the literature to arrive at best estimates of the extinction budget.

Empirical Orthogonal Function Analysis

Empirical orthogonal function (EOF) and possibly other types of eigenvector analysis will be done to help summarize the data and gain insights into possible physical mechanisms at work. When working with large amounts of data, EOF analysis is especially useful by effectively reducing the dimensionality of the data set. A large number of observations at many locations can be reduced into a reasonable number of spatial patterns (eigenvectors), with a time series associated

with each eigenvector showing the time variability of each pattern.

The EOF analysis is purely a statistical technique that attempts to account for most of the variability in a data set by a few eigenvectors. Although no physics is explicitly included in the analysis, the data set represented by the eigenvectors is certainly affected by physical processes. EOF analysis in conjunction with sound physical reasoning, including knowledge of meteorological conditions, location of emission sources, etc. can help in the formation of hypotheses and provide a qualitative check of receptor and deterministic modeling results.

The variables for which EOF analysis is likely to be done are sulfate, SO_2, tracer, elemental carbon, organic carbon, fine soil, and certain trace elements. EOF analysis of the vector wind field may be done as well. EOF analyses of the modeled output wind and concentration fields will be investigated as a method to help organize the large amount of model output. Additional EOF analysis using two (or more) parameters such as sulfate and tracer can be used to identify common jointly occurring patterns of more than one parameter.

Meteorological Classification

A meteorological classification scheme will be developed and applied to the study years and several previous years. The scheme will classify days into types on the basis of similarity of meteorological parameters. There are several reasons for doing a classification. One reason is to compare the frequency of each weather pattern during the study year with other years to determine how representative the study year is. Each pattern is likely to have transport of visibility affecting pollutants from different areas; the relative frequency of patterns for the study year compared to long-term averages can help put the impacts during the study year into perspective. It also provides a logical method of stratifying the data of the study year into a manageable number of patterns. Averaged spatial patterns of sulfate, etc. along with the variation within each pattern can reveal the main pathways for transport of both hazy and clear air into the study area. Contributions from individual source areas may also be inferred from the concentration fields associated with each pattern.

The meteorological classification scheme can aid in the interpretation of the EOF analyses. The time series of the EOF analyses indicate the times a particular eigenvector is significant. By determining the corresponding meteorological pattern most commonly associated with each eigenvector, it is easier to interpret the physical factors associated with the eigenvectors.

The classification scheme will also be used to study the MPP outage of June-December 1985. Sulfate concentration levels and spatial patterns associated with each weather pattern will be compared for the outage year and other years with SCENES data. This will help put bounds on the contribution of MPP to regional sulfate levels.

Of critical importance in the classification scheme are surface and upper

air wind speed and direction, atmospheric moisture and thermal stratification. The wind data are necessary to account for the transport and dispersion properties of the flow. Moisture is necessary to determine the potential for aqueous phase oxidation of SO_2 to sulfate and washout. Thermal stratification is needed to know if pollutant emissions are likely to remain trapped in basins or are mixed through a deeper layer of the atmosphere.

11. Attribution

Overview

The attribution analysis will be done using a variety of analytical tools; particularly deterministic, hybrid, and receptor models; and the MPP emission modulation study. Deterministic modeling is an approach that attempts to explicitly account for physical and chemical processes transporting and acting on emissions from a source. Receptor models use measurements made at the area of concern (receptors) along with characterization of the emissions from sources potentially affecting the receptors. The contribution of each source to concentrations at the receptors is determined statistically through multivariate analysis techniques which link the sources to the measured concentrations. Hybrid models use a combination of deterministic and receptor modeling techniques. Apportionment implies determining the concentration of sulfate at the receptor areas resulting from MPP and other sources. The apportionment of secondary aerosols such as sulfate is a complex problem as noted by the National Research Council (1990) and others. Transport, dispersion, deposition, and transformation must be accounted for.

Results from the extinction budget analysis will be used in conjunction with the sulfate attribution to determine the fractional contribution of MPP sulfate to the extinction coefficient. The effect of primary particles will also be considered. The next step is to evaluate the perceptibility of the contribution of MPP to the extinction coefficient. Finally, the question of the effect of reducing emissions from MPP upon visibility will be addressed. Each of the main study components used in the attribution analysis is described in the following sub-sections.

The results from the various models and analyses will be compared and reconciled. Reconciliation is a critical component of the analysis. If results from a particular model or analysis cannot be reconciled with other analyses, the results will not be used. The range of uncertainty in each calculation and the reconciled results or consensus will be estimated.

A major area of concern that has been expressed by some is the possibility for misuse of tracer data for source apportionment. Specifically, two issues have been expressed: (1) that any tracer level above background measured at the receptor sites will be interpreted as attribution of visibility impairment by tracer sources; (2) that tracer data will be incorporated into analyses inappropriately by repeated regression analysis with whatever parameters and formulisms are needed until a statistically significant relationship is found, though no physical relationship is evident.

Project MOHAVE planners do not interpret the appearance of any tracer above background as sufficient criteria to indicate visibility impairment and will contest any who make such a claim. To demonstrate good faith and concern for appropriate scientific methods, Project MOHAVE

will arrange for tracer data to be withheld from all who have any role in attribution analysis until such time as physically meaningful empirical source attribution formulisms have been developed based upon other Project MOHAVE data. This will be done to promote the development of physically reasonable models prior to the availability of tracer data, and to avoid the appearance of forcing the models to fit preconceived notions.

Deterministic Meteorological Modeling

Deterministic meteorological modeling is based on fundamental physical conservation relationships. These relationships include conservation equations for momentum, temperature, mass, and the three phases of water. Meteorological modeling for Project MOHAVE will be done by Colorado State University using the Regional Atmospheric Modeling System (RAMS). A brief overview of RAMS is given in Appendix 6. Additional information about RAMS appears in Pielke *et al.*, (1990). A dedicated super workstation (IBM RISC) will be used for the modeling. The model will provide detailed wind and turbulence fields and a prediction of cloud height and location. Cloud predictions will be checked against satellite photographs.

The meteorological domain for the simulations will cover the southwestern United States. To obtain better terrain resolution near MPP, a telescoping nested grid will be used. In a nested grid approach, the larger scale results provide the boundary conditions for input into a finer scale modeling domain. The entire one year study period will be modeled. For selected cases from the intensive study periods, modeling with much finer resolution will be done. The preliminary grids to be used for the analysis are shown in Figure 10. Grids 1 and 2 will be used for the year-long study; the case studies will also use grids 3, 4, and 5. The horizontal and vertical number of grid points, and the horizontal grid spacing for each grid are shown below.

Grid	# of grid points	Spacing (km)
Grid 1	x=100 y= 60 z=44	32
Grid 2	x=104 y= 72 z=44	8
Grid 3	x=144 y=144 z=44	2
Grid 4	x= 80 y=80 z=44	0.5
Grid 5	x= 80 y=80 z=44	0.5

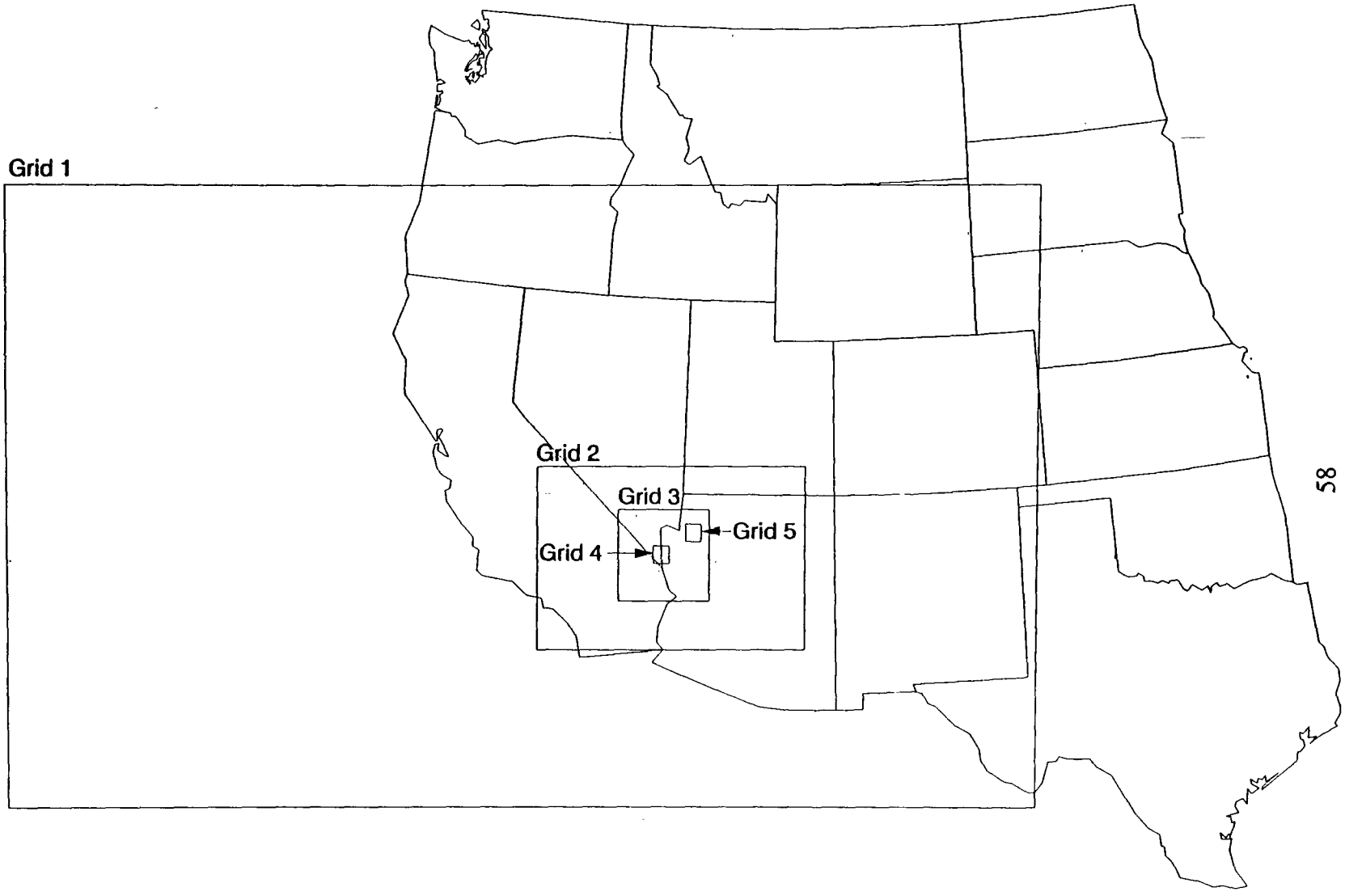

Figure 10. Meteorological modeling grid.

The model is initialized every 12 hours using the analysis field supplied the National Meteorological Center's Nested Grid Model. The Nested Grid Model uses surface and upper air data to generate initial fields of variables such as pressure, temperature, moisture and wind. The CSU RAMS modeling takes this initial field and generates mesoscale fields for the next 12 hours before being re-initialized.

The use of data from the wind profilers will be investigated for use in four-dimensional data assimilation. In this mode, the measured data are used to adjust, or "nudge" the model results. Data from the profiler site at Truxton are the most likely to be used for nudging. The Truxton site is in relatively open terrain and more likely to be representative of general flow in the study area than the other sites which are expected to be considerably influenced by local terrain features. Data from the radar wind profilers not used in the nudging process will be used to evaluate the performance of the model. The evaluation will be done for every hour having modeled and wind profiler data. A quantitative comparison between the predicted and observed winds will results in "figures of merit" for model predictions as a function of meteorological conditions. The wind and turbulence fields obtained from the deterministic meteorological model will provide the necessary input to calculate the transport and dispersion of MPP and other emissions of interest.

Transport, Chemical and Deposition Modeling

Once the wind fields have been determined, a model is needed to account for transport, chemical transformation and deposition. The transport model to be used is a version of the CAPITA Monte Carlo model currently being developed for EPA under a cooperative agreement with Washington University in St. Louis. A copy of the cooperative agreement proposal, which describes the modeling approach in more detail, appears as Appendix 7. The model is being developed specifically for visibility related studies. Evaluation and calibration of the model is being done using data sets such as IMPROVE, SCENES and NESCAUM. Modifications to the model to fully utilize the wind, turbulence, and moisture field supplied by the meteorological model may be necessary.

In the modeling approach, simulated pollutant quanta (particles) are "emitted" from each source. These quanta are moved in fixed time increments using wind fields supplied by the meteorological model. During transport the pollutant quanta are subject to chemical transformation and removal. The dispersion is achieved by imposing a randomized perturbation to the trajectory at each time step. Transformation and removal are also imposed as stochastic events at each time step. The result of the Monte Carlo simulation is a large number (10^5-10^6) of pollutant "particles" dispersed geographically for every time step of the simulation. The model is considered a Monte Carlo simulation because of the probabilistic treatment of transport, transformation, and removal.

The model will make use of the turbulence field generated by the meteorological model to perturb the trajectory. The moisture fields given from the meteorological model will be used to select between wet (heterogeneous) and dry (homogeneous) conversion rates in the SO_2 to sulfate transformation parameterization. Details of the modeling methodology are still to be determined. The model is based on that described by Patterson *et al.* (1981).

Hybrid and Receptor Modeling

Measurements of endemic and artificial tracers will be used to estimate the transport and dispersion of MPP and other sources. The transport can be verified by checking with trajectories given by the meteorological model. The transformation and deposition of SO_2 and sulfate are also necessary; these may be parameterized based upon such information as solar radiation, moisture (especially clouds), oxidant availability, and vertical mixing. The hybrid models will use tracer measurements to account for transport and dispersion, and parameterizations to account for deposition and transformation.

Versions of the chemical mass balance (CMB), differential mass balance (DMB), and possibly the tracer mass balance regression (TMBR) models will be used. CMB uses the relative ratios of natural or man-made tracers at the sources and receptor locations to apportion primary species for each measurement period. CMB will be used to apportion primary species using the high volume dichotomous sampler data. DMB, a hybrid model, uses trace material to establish dispersion factors and calculates the effects of deposition and oxidation. TMBR uses the variation of trace material over time to estimate primary or secondary aerosol contributions from each source. In its original formulation, TMBR requires a constant tracer to SO_2 emission ratio. However, Project MOHAVE will use a tracer emission rate that will be only approximately proportional to SO_2 emissions. TMBR may be used in an exploratory mode to investigate the effect of departure from model assumptions. Any use of the results must acknowledge and quantify the effects of departure from the model assumptions.

In DMB and TMBR it is assumed that each source has a uniquely emitted tracer associated with it. The best available emissions and source characterization data will be used to identify unique tracers for each significant source. If unique tracers are not available, CMB may be applied first to partition the ambient species concentrations into components attributable to the various groups of sources.

The NAS review of WHITEX noted a number of concerns about the use of DMB and TMBR. A summary of the NAS WHITEX comments and the steps that will be taken by Project MOHAVE to help resolve these issues appears as Appendix 8. The DMB and TMBR models will be modified to help alleviate concerns in the NAS review of WHITEX by taking advantage of the more detailed meteorological fields generated by the meteorological modeling that is a part of Project MOHAVE. For example, the effect of moisture upon conversion

rates for both DMB and TMBR will utilize the moisture fields generated by the meteorological model.

As mentioned above, the implementation of DMB will differ from WHITEX by incorporating physical processes in a more robust manner. In Project MOHAVE, the effect of moisture on sulfate formation will be treated more rigorously than in the WHITEX study. In addition to surface moisture measurements, the deterministic meteorological model will give calculations of moisture at many vertical levels. This information will include prediction of clouds, which can be compared to satellite photos and surface observations. Rather than scaling linearly with surface relative humidity, the probability of plume-cloud interaction will be estimated and used to assign an SO_2-to-sulfate conversion rate. Rates of sulfate formation occur rapidly in clouds, and much slower without clouds. Different conversion rates based upon whether or not clouds are present should more appropriately account for the effect of moisture on conversion rates.

In DMB, as currently formulated, deposition and conversion rates are constant; the assumed rates are multiplied by plume age to give sulfate concentrations and deposition loss. Trajectory calculations are used to give plume age. Dispersion is accounted for by ratioing ambient trace material concentrations attributable to a source by known trace material release rates. In Project MOHAVE variable conversion rates will be used, such as described above. With the deterministic meteorological modeling wind fields, reliable plume age calculations should be possible.

The equations for, and assumptions used in, CMB, DMB, and TMBR as presently formulated are given in Appendix 9.

Extrapolation of Intensive Study Periods to the Long-Term

To determine longer term impacts to visibility at GCNP, it is necessary to extrapolate from results of the intensive study periods. This will be a two-step process; the first step will relate the entire 12 month study period to the intensive period, while the second will extrapolate from the 12 month period to a multi-year period. The first step involves application of source-oriented and hybrid models, which will be developed and evaluated with intensive period data, to the meteorology and air quality data for the entire study period. In the second step the relative frequency of long-term meteorological patterns will be compared with those of the study period.

Deterministic models will be evaluated and calibrated using the more complete data of the intensive periods. The resulting models will then be run with data from the entire study period. For all modeling analyses a portion of the data may be withheld in order to independently test the models.

During the intensive study periods, hybrid modeling will use the artificial tracer results for MPP and any other sources tagged with artificial tracers. Hybrid models will also use endemic tracers for the remaining significant sources.

Results from hybrid models based on endemic tracers will be compared to results of the same models using artificial tracers to evaluate the utility of endemic tracers. If successful, models using endemic tracers will then be applied to the entire study period (covering a complete annual cycle) and used in conjunction with the deterministic modeling analysis.

The representativeness of the study year to longer term average conditions will be studied. It should be acknowledged that significant year to year variability in meteorological conditions occurs and that the likelihood of any given year being "typical" is not high. The frequency of occurrence of each meteorological regime identified in the meteorological classification process described in Section 11 will be compared for the study year and other years for which data are available. Where they exist, optical and air quality measurements from previous years will be compared to the study year measurements. The frequency of occurrence of each pattern for the study period and longer term average can then be compared to put the study year into perspective.

MPP Emission Modulation Study

The MPP was inoperative for a seven month period from July to December 1985. This presents a unique opportunity for investigating the effects of MPP. The MPP emission modulation study, discussed in Section 2 and Appendix 5 is a potentially powerful receptor approach to estimate the extent of MPP contributions to downwind sulfate levels. The analysis will be conducted by using a meteorological classification scheme to control for year-to-year variations in meteorology, and comparing measured sulfate at Spirit Mountain, Meadview and Hopi Point for periods of varying MPP and other SO_2 emissions. The study will include the following elements:

Independent statistical analysis of the experiment.

Chemical analysis of all filters. (Quality assurance will be evaluated through comparison of current results to past data through regression and time series analysis).

Classification of the synoptic and mesoscale weather patterns (meteorological regimes) affecting transport of the MPP plume.

Deterministic wind field, transport, and dispersion modeling for each of the meteorological regimes.

A detailed compilation of regional SO_2 emissions data for the control and outage period to allow an accounting for variation in SO_2 emission patterns.

All data manipulation will be performed in the "blind" to avoid charges of bias or data selection. Results of the study will be used along with the modeling and other analyses to estimate the effect of MPP on visibility at GCNP.

Framework for Interpreting Results

In a complex program such as this, a sound plan for compilation of results is as important as the collection of high quality and representative data and the performance of appropriate interpretive analysis. Development of an approach to organize the results from this program helps to focus attention and resources on critical steps for the entire program and communicate those ideas to others.

Just as it is inappropriate for worst case results to receive primary attention, it is also inappropriate to dwell on average or typical conditions, especially for an instantaneous effect such as visibility. The 12 month study period with hourly deterministic model results requires some method for summarizing the results of the study that avoids these pitfalls. A preliminary conceptual framework for summarizing the results of Project MOHAVE is shown in the table on the following page. The key idea is the stratification of time periods based upon
the locations with respect to GCNP of MPP emissions and those of other significant sources, such as from southern California. These would be based upon the modeling studies. Another stratification is whether the plumes have undergone wet or dry chemistry (based upon modeling results and observations). If useful, other stratifications could be developed. The frequency of each condition, the average and standard deviation of the percent sulfate from MPP, the percent of extinction from MPP and a measure of the perceptibility of the MPP impact is estimated for the study period.

Extrapolation to a long-term average will be done through the use of a meteorological classification scheme as previously described. This type of approach provides an efficient manner of presenting the magnitude and frequency of estimated MPP emissions on GCNP over a long-term period that could be used to evaluate the significance of existing impairment.

Conceptual Framework for Summarizing Project MOHAVE Results

GCNP Impact & Condition	Frequency (Deterministic Model)	% Sulfate (Reconciled Models)	% Extinction (Extinction Budget)	Measure of Perception
No MPP in GCNP				
MPP & SCA Dry				
MPP & SCA Wet				
MPP & Other Sources Dry				
MPP & Other Sources Wet				
MPP Alone Dry				
MPP Alone Wet				
Other Appropriate Categories				

SCA refers to the urban and industrial areas of southern California.

12. Overall Quality Assurance

Approach

An independent quality assurance audit will be done by ENSR. The major emphasis of independent quality assurance in Project MOHAVE will be upon verifying the adequacy of the participants' measurement procedures and quality control procedures, and upon identifying problems and making them known to project management. Although routine audits will play a role, major emphasis will be placed upon the efforts of senior scientists in examining methods and procedures in depth. This approach will be followed because fatal flaws in experiments emerge not from incorrect application of procedures by operators at individual sites or laboratories, but rather from incomplete procedures, inadequately tested methods, deficient quality control tests, or insufficient follow-up of problems.

System Audits - Study Planning and Preparation

Senior auditors will review study design documents to ensure that all measurements are being planned to produce data with known precision and accuracy. The auditors will verify that adequate communications exist between measurement and data analysis groups to ensure that measurements will meet data analysis requirements for precision, accuracy, detection limits, and temporal resolution. Quality control components of the measurements will include:

Determination of baseline or background concentrations and their variability.

Tests for sampler contamination.

Adequate and precise measurement of aerosol and tracer sampler volume and time.

Blank, replicate, and collocated samples.

Assessment of lower quantifiable limits (LQL), and determination of measurement uncertainty at or near the LQL.

Regular calibrations and calibration checks, traceable to standard reference materials.

Procedures for collecting QC test data and for calculating and reporting precision and accuracy.

Periodic QC summary reports by each participant.

Documented data validation procedures.

Verification of comparability among groups performing similar measurements.

A senior auditor will visit each measurement group, laboratory and data management and analysis group prior to the intensive field studies to verify that adequate progress is being made toward beginning measurements on schedule and within acceptable quality limits. A thorough review of written procedures will be part of this evaluation, including a review of all standard operating procedures. Issues to be addressed include:

Availability of equipment and supplies.

Manpower availability.

Readiness of written procedures and data collection protocols.

Adequate sample ID and sample tracking system.

Thoroughness of method evaluation tests.

Understanding of QC procedures and adequacy of protocols for collecting QC test data.

Testing of software used for data management, data validation, and data analysis.

Measurement System and Performance Audits

Audits of the field sites, the laboratories, and the data management and analysis center will be conducted once during the study, probably at the beginning of the winter intensive measurement period. System audits will verify that the items described in the system audits section are being applied. Performance audits will include:

Field sites - Instrument calibration checks, leak checks on aerosol and tracer samplers, and on the tracer injection system.

Laboratories - Relabeling of existing samples by ENSR and reanalysis by the study laboratories to verify precision and reproducibility. Submittal of prepared samples of known concentration, where needed. If the

laboratory already participates in a regular intercomparison program or if it uses standards directly traceable to NIST, then a system audit will verify this, and no additional samples will be prepared.

Data management - Manual calculation of derived concentrations and uncertainties.

Data analysis - Manual data traceability tests to verify pre-analysis processing.

Based on audit results and discussions with project management, the auditors will identify problems which have the potential to jeopardize data quality. They will provide immediate feedback to operational personnel and will provide letter reports following the audits. Corrective action request forms, to be completed by operational personnel and returned to the auditor, will verify that problems have been addressed. Throughout the study, the auditors will review the participants' QC summary reports.

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Appendix 1

Project MOHAVE Update Summary - September 16, 1991

Study Component	Description of Study Component	Responsible Party
Schedule	Overall - 12 months starting in Sept. '91 Intensives - Two 4 to 6 week intensives, (1) January '92, (2) July and August '92.	
Emissions	Conduct review of available emission data and compile inventory. Do source profiling of MPP during a portion of each intensive period. *Continuous source sampling at MPP: SO_2, NO_x and particulate concentrations (plus frequent particle composition).**	Bruce Polkowsky, OAQPS

* Italics used to indicate unfunded study components.

Study Component	Description of Study Component	Responsible Party
Deterministic Modeling	Deterministic meteorological modeling (wind, turbulence and moisture fields) for each day of the 12 month period with domain and resolution as feasible.	Roger Pielke, CSU
	Apply a Lagrangian Monte Carlo transport model with some chemistry included to transport, disperse and chemically transform the plume using the output from the meteorological model.	William Wilson, RTP
	Full chemistry deterministic modeling (RADM, with enhanced particle treatment, in-cloud processes and optics) for about 20 to 30 selected days. Expected input includes ammonia gas, particulate ammonium ion, and hydrogen peroxide measurements at a few locations during intensives. Would employ the output from Pielke's modeling.	*Jason Ching, NOAA-RTP*
Tracer	Continuous in-stack release of perfluorocarbon tracer during each of two intensive periods. Sampling at 31 sites on 12-hour (at 4 receptor sites) and 24-hour (all other sites) sampling schedule (see attached map for sampling locations). The Hopi Point and Meadview sites will each have two additional collocated samplers. A 21 day pre-release sampling pre-test at all sites to establish background levels and for QA.	Russell Dietz (overall), Brookhaven Nat'l Lab and Ray Dickson (tracer release), NOAA Idaho Falls
	*Additional perfluorocarbon tracers released to tag the Los Angeles Basin and San Joaquin Valley.**	*Dietz & Dickson*
	Increased time resolution of tracer data: 6 hours at receptor sites, 12 hours at other sites.	*Dietz & Dickson*

Italics used to indicate unfunded study components.

Study Component	Description of Study Component	Responsible Party
Meteorologic monitoring	12 month period: Continuous vertical wind profiling over the 12 month period at MPP plant site and Truxton using radar wind profilers. A Radio Acoustic Sounding System (RASS) will also be deployed at the plant site to give boundary layer vertical temperature structure. A doppler sodar will be deployed at Meadview most of the study period to measure wind profiles. Additional instrumentation to include at least 4 surface meteorology stations with pressure sensors (temperature & relative humidity also) to examine response of locally channeled flow to larger scale pressure gradients (November 1991-August 1992).	John Gaynor, NOAA Boulder
	Surface meteorological stations at the 4 receptor sites measuring wind speed, wind direction, temperature, relative humidity and solar radiation.	John Molenar, Air Resource Specialists
	Intensives: Two additional radar wind profilers will be operated during the winter intensive near Needles and Temple Bar. Two additional profilers during the summer intensive will be located at Meadview and Cottonwood Cove. The Meadview wind profiler will replace the sodar, which will be moved to Temple Bar for the summer intensive. Radar wind profiler data from Los Angeles Basin and western Mojave Desert sites will also be available for the summer intensive. Surface meteorological stations at radar wind profiler sites measuring winds, temperature and relative humidity. Some special studies using tethersondes and/or radiosondes may be done at locations of interest.	John Gaynor

Study Component	Description of Study Component	Responsible Party
Air quality monitoring	Particle Monitoring: Full IMPROVE samplers at the 4 receptor, 4 IMPROVE sites and 2 Improve protocol sites.* IMPROVE channel A at all 21 remaining sites. See attached map for site names and locations (total of 31 sites). Drum sampling in 8 size ranges at the receptor and 2 additional sites, with 4 or 6 hour resolution. Selected drum sampler filters to be analyzed. Intensives: two 12 hour samples per day, every day at receptor sites; 24 hour samples every day at all other sites. Tracer and SO_2 sampling at all sites following the particle sampling schedule. H_2O_2, NH_4 and NH_3 monitoring periodically during intensives. Hi vol dichotomous samplers and annular denuder samplers at 3 sites for high sensitivity particulate analysis necessary for CMB modeling. More details will be available soon. Remainder of study year: 24 hour particle and SO_2 sampling with IMPROVE samplers every Wednesday and Saturday at receptor, IMPROVE and IMPROVE protocol sites. *Increase number of particle monitoring sites for non-intensive periods.***	Bob Eldred, UC-Davis

* *The IMPROVE sampler has 4 channels. Channel A collects fine particles (<2.5μm) on a teflon filter and provides total fine mass, elemental analysis (H and Na-Pb), organic and elemental carbon and absorption. Channel B uses a fine nylon filter for ions (Cl^-, NO_2^-, NO_3^- and SO_4^{2-}). Channel C is used to obtain organic and elemental carbon from a fine quartz filter. Channel D measures PM-10 total mass on a teflon filter and SO_2 with an impregnated quartz filter.*

Study Component	Description of Study Component	Responsible Party
Optical monitoring	Continuous monitoring for the entire period. Nephelometers at all receptor sites and a transmissometer added at Meadview, in addition to ones already at IMPROVE sites.	John Molenar, Air Resource Specialists
	*Airborne lidar aerosol mapping several weeks during the intensive periods.**	*Jim McElroy, EMSL-Las Vegas*
Data Interpretation	Analysis of historic meteorologic data to optimize timing of intensive periods. Analysis of MPP emission modulation (1985 shut-down).	Mark Green, DRI
	Eigenvector Analysis	Mark Green
	DMB modeling	Bill Malm, NPS
	CMB modeling with high sensitivity particulate data.	Robert Stevens, RTP
	Extinction Budget	Marc Pitchford, EMSL- Las Vegas
	Reconciliation of results from receptor & deterministic modeling and eigenvector analysis, extinction budget, trajectory analyses, etc. Overall summary of results.	Marc Pitchford

** *Italics used to indicate unfunded study components.*

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Study Component	Description of Study Component	Responsible Party
Quality Assurance	Each component of the study is responsible for QA on its' portion of the study. Overall QA audit covering all portions of the study to be done by independent reviewer.	 Charles McDade

Appendix 2

Project MOHAVE[1] Conceptual Plan

Introduction

This plan documents the thoughts and intentions of those who are preparing to determine the contributions by the Mohave Power Project (MPP) to haze in Grand Canyon National Park (GCNP). Its purpose is to provide a vehicle to obtain review and comment by various interested parties at an early point in the planning process when adjustments are more easily accommodated. This conceptual plan is designed to provide overall guidance to the technical experts who are responsible for developing the more detailed study plan.

The first part of this paper contains information on the study background, objectives, and an overview of the approach. This is followed, in the second part, by an expanded discussion of the approach which contains information on the visibility attribution process, use of artificial tracers, ambient monitoring, and data interpretation and models.

Background

The 1991 fiscal year budget for the United States Environmental Protection Agency (EPA) includes a Congressional "add-on" at the level of $2.5 million for a 2-year effort titled "Pollution tracer study at the Mohave Powerplant". Discussion has revealed that congressional intent was to have EPA perform a study to assess MPP's contribution to visibility impairment in GCNP. Members of congress have demonstrated an interest in visibility impairment in the Federal Class I Areas (i.e., national parks and wilderness areas meeting certain requirements); and in particular an interest in GCNP impairment by large point sources of SO_2. For many this interest was intensified by the results of the 1987 Winter Haze Intensive Tracer Experiment (WHITEX) conducted by the National Park Service (NPS).

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[1] While Mohave is the name of a coal-fired power plant in Nevada, Project MOHAVE contains an acronym for Measurement Of Haze and Visual Effects.

WHITEX involved a six-week long intensive monitoring study during which an artificial tracer was released from the Navajo Generating Station (NGS)[2]. NPS analysis of optical, air quality, and meteorological data indicated that a significant fraction of the winter hazy periods in GCNP were due in large part to sulfates resulting from NGS emissions. EPA used these results as the basis for proposing additional emission controls at NGS. The WHITEX data analysis methodology, results, and use of the results were cause for considerable controversy.

In an attempt to resolve the technical issues raised by WHITEX, the National Research Council of the National Academy of Sciences (NAS) was requested to consider the relative importance of human derived and natural emissions that contribute to visibility reduction. The Council established a Committee on Haze in National Parks and Wilderness Areas. One task of the committee was to evaluate WHITEX. Their report neither wholly endorsed nor discredited the NPS WHITEX findings, though it did provide an illuminating discussion of the technical issues. In an effort to avoid some of the controversy of WHITEX and to take advantage of the expertise assembled by NAS, Project MOHAVE has requested the opportunity to discuss this conceptual plan with the committee. The committee is scheduled to be briefed on this effort in early Spring 1991.

Salt River Project (SRP), the operators of the NGS, in an attempt to resolve their doubts concerning WHITEX, supported a more extensive tracer study in the winter of 1990. Though only preliminary results of this study are now available, it appears to also indicate NGS emissions in GCNP during haze, though at a lower frequency of occurrence.

It is the goal of the planners of Project MOHAVE to take advantage of the best and most successful aspects of the WHITEX and SRP studies, and to address the issues raised by the NAS WHITEX review to the maximum extent possible, and to use and extend information previously obtained by numerous efforts.

Previous air quality studies in the region containing the desert southwest (including SCENES, VIEW, VISTA, WRAQ and RESOLVE) provide a great deal of background information useful to the planning of this project. Prevailing

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[2] NGS is a 2250 Mw(e) coal-fired powerplant located near Page, Arizona, approximately 25 Km northeast of GCNP.

southwest winds, especially in the summer, carry MPP emissions toward GCNP. They also carry emissions from the southern California urban/industrial area towards GCNP. There is considerable evidence that southern California is the dominant source area of pollutant haze for GCNP. A major technical challenge for Project MOHAVE is to separate the influence of MPP from that of southern California and other regional influences.

The most important man-made pollutant species responsible for GCNP haze are particulate sulfates. These are generally formed in the atmosphere by chemical conversion of gaseous SO_2, which is emitted by combustion of fuel containing sulfur. Other particulate components important to GCNP haze, organics and crustal species, are from natural and man-made sources. GCNP visibility levels are often so good that light scattering by air molecules (Rayleigh scattering) is also a significant contributor to the extinction coefficient.

MPP's most significant potential contribution to GCNP haze is by emissions of SO_2 that are converted to sulfates. Other sources contributing to particulate sulfate are southern California (primarily by oil refineries), other coal-fired power plants (e.g., Reid Gardner north of Las Vegas, NV and NGS near Page, AZ), copper smelters in southern Arizona, New Mexico, northern Mexico and Utah and oil refineries in Texas and the Monterrey area of Mexico. Other sources which may influence GCNP visibility are large urban areas (e.g., Las Vegas, NV, Phoenix/Tucson, AZ and the Wasatch Front in Utah) and wildfires. These sources are expected to be more dominated by organic and elemental carbon pollutants than sulfate.

Objectives

This conceptual plan considers two related objectives: (1) to determine the MPP contribution to GCNP haze and (2) to determine the relative contributions of the major pollution emission sources (including MPP) affecting GCNP haze. For both objectives, determining the contribution to GCNP haze implies a quantitative evaluation of intensity, spatial extent, frequency, and duration. The intensity of haze contributed by a source includes both an absolute physical measure of haze (e.g., contribution to the extinction coefficient) and its perceptibility (e.g., scenic element contrast change, or change in modulation transfer function). A part of both objectives is an assessment of the changes in visibility at GCNP that would be expected if MPP emissions were changed.

The first objective implies determining the contributions to GCNP haze by two source categories: MPP and a composite of all non-MPP sources. The second objective expands upon the first objective. Instead of concentrating on one source's impact, it calls for simultaneous assessment of all the important sources of haze for GCNP. There is no doubt that a study designed to meet the first objective would also address other sources to some extent. However, this would be incidental to the first objective, unlike the second objective where it is the primary focus.

A program designed to meet the second objective is beyond the resources presently available for this effort. Unless additional support becomes available Project MOHAVE will be designed to meet the first objective and to prepare a foundation for further investigation of the impact of regional haze in GCNP.

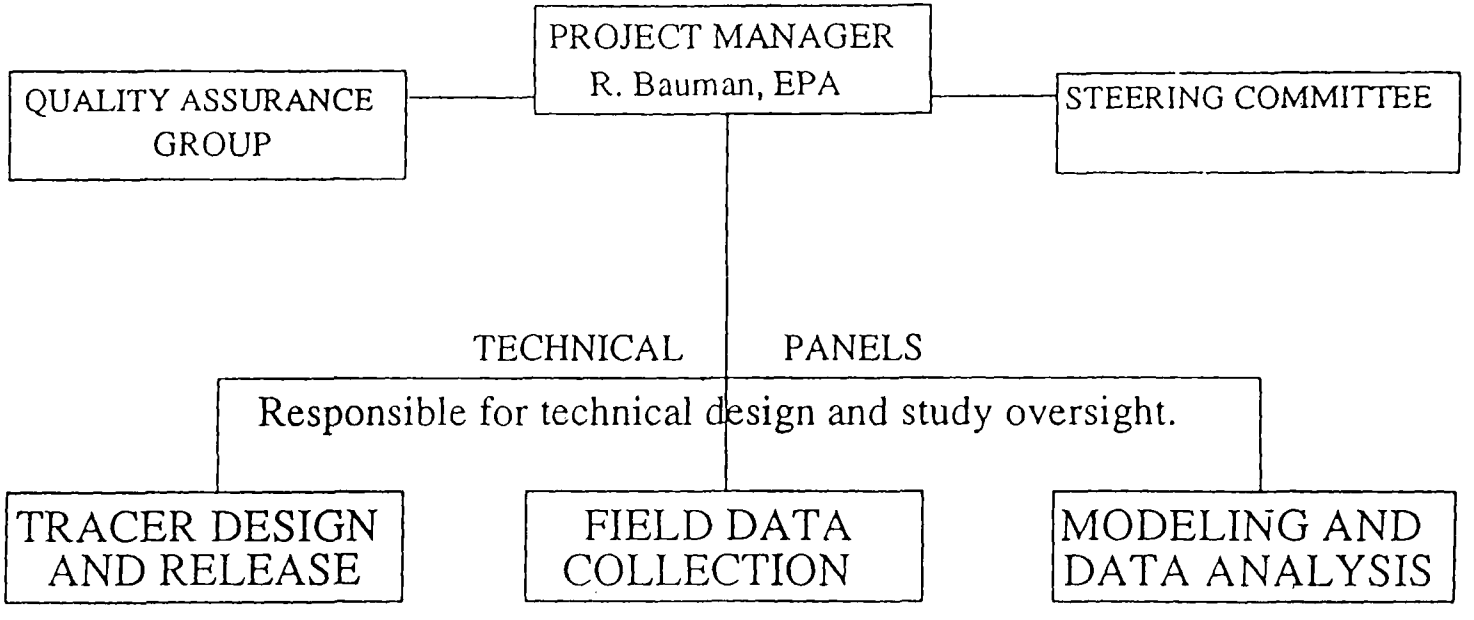
Approach Overview

The EPA Office of Air Quality Planning and Standards (OAQPS) has overall management responsibility for Project MOHAVE. Robert Bauman, the OAQPS Project Leader, has selected a Project Steering Committee to advise him on the overall direction of the study. Several technical advisory panels are being constituted to provide recommendations at a greater level of detail. Experts selected for the technical advisory panels will provide the primary means for Project MOHAVE to incorporate insights gleaned from earlier investigations. Figure 1 indicates the program's management/advisory structure.

Project MOHAVE will use sophisticated deterministic and receptor modeling to identify the MPP influence. During two intensive study periods (four to six weeks each), a unique tracer material will be combined with the MPP emissions in concentrations sufficient to be detected hundreds of kilometers away. The tracer will provide a check of the deterministic modeling results and provides a unique signature for the MPP plume, for use in receptor modeling. The main emphasis will be on deterministic modeling, with secondary emphasis given to receptor modeling. It is not prudent to implicitly trust the results of either modeling approach alone; thus both approaches will be tried. If results of the two approaches are substantially different, an in-depth investigation into the reasons for the differences and an evaluation of the results will be done before any conclusions are reached regarding MPP's impact.

The intensive periods will be selected to optimize the chances of establishing the maximum contribution to GCNP impairment by MPP.

PROJECT MOHAVE



- Tracer selection
- Release mechanisms
- In-stack monitoring
- Release protocol

- Equipment selection
- Site selection
- Ambient monitoring
- Optical monitoring
- Meteorology
- Data processing
- Filter analysis

- Dispersion modeling
- Trajectory analysis
- Receptor modeling
- Extinction budget
- Attribution analysis
- Perception & effects

2-4a

Tentatively these would be the summer monsoon season and the mid-winter storm season. Both periods have the possibility of transporting MPP emissions to GCNP decoupled from southern California emissions, and sufficient moisture for possible liquid phase conversion of SO_2 to sulfate (much faster conversion than the alternative gas phase reactions). These conditions are intermittent even during the periods of their greatest frequency. Thus the intensive periods will also include more typical summer and winter conditions where MPP influence in GCNP is not expected to be as great.

To further ensure that data from the intensive periods can be interpreted in terms of longer term typical conditions, the overall study period will be 12 to 15 months. During the non-intensive periods of the study, air quality and meteorology measurements will be made at numerous locations throughout the study area. Intensive study period data will be used to evaluate source-oriented deterministic models using augmented upper air meteorological data and receptor models based upon endemic tracers. The deterministic models will then be applied to the entire study period. If successful, receptor models using endemic tracers will be also be applied using data collected for the entire study period. Finally, the study period results will be extrapolated to the long-term by comparison with and if necessary adjustment to climatological characteristics of importance.

A tentative schedule for Project MOHAVE calls for field measurements to start in July 1991 and continue until September of 1992. The winter intensive period will be in January 1992, with the summer intensive period from mid-July to late August 1992. Data interpretation and report preparation is anticipated to continue for approximately 18 months after the end of the field monitoring program.

Approach

The attribution of impacts from MPP and other sources will ultimately be derived from an extinction budget by air pollutant species. The majority of the MPP impact is expected to be from secondary sulfate particles. Measurements of the particle components such as sulfate, nitrates, carbon and crustal species are related to optical measurements by statistical and first principle approaches to produce the extinction budget.

It is expected that the contribution of sulfate particles from MPP and other sources will be estimated primarily from deterministic modeling. Receptor

modeling will also be done for this purpose, providing a check of the deterministic modeling analyses. The results for the two types of models will be compared for consistency (model reconciliation). Eigenvector analysis will also be done to support results from the modeling studies. During the two intensive periods, an artificial tracer will be injected into the MPP plume. The tracer provides a check of the transport and dispersion calculated by the deterministic modeling. It also provides a unique signature of the MPP plume for use in receptor modeling. To estimate impacts for the remainder of the study period, deterministic modeling will be performed and receptor modeling using endemic tracers will be investigated.

Substantial monitoring will be required to support the extinction budget and modeling studies. This will include ground based and airborne meteorologic, air quality and optical measurements and remote sensing of vertical wind and temperature profiles. Expanded descriptions of the main components of the study follow.

Tracer

During the intensive study periods an artificial tracer will be released continuously either through the stack at MPP or by balloon at plume height in the immediate vicinity of the power plant. A stack release would give more confidence that the plume and tracer are well mixed and is the preferred method. However balloon release of tracers has been routinely done (NOAA, Idaho Falls) and is a feasible alternative. For objective 2, different artificial tracers would be released at other sources or source areas to tag their emissions more precisely than through the use of endemic tracers. Other sources to tag may include the San Joaquin Valley (Tehachapi Pass), the Los Angeles Basin (Cajon Pass), Las Vegas, Reid Gardner Powerplant, Navajo Powerplant and copper smelters.

Tracer can be released at a constant emission rate or at a constant ratio of tracer to SO_2. Variation of tracer to SO_2 ratios was a complicating factor in the WHITEX receptor modeling analysis. If released at a constant rate, SO_2 emission rate variations would complicate the receptor modeling, requiring adjustment of the ratio of tracer to sulfur dioxide concentration. This requires knowledge of plume age. However, for use in deterministic modeling, it is more desirable to have a constant tracer emission rate, to simplify the dispersion calculations. Also, the deterministic model can give the plume age necessary to adjust the tracer to sulfur dioxide emission rates in the receptor modeling.

Ideally, a tracer should closely mimic the species of interest for receptor modeling and chemical transformations; in this instance SO_2 and its conversion

to SO_4 and deposition of the sulfate particles. This would suggest using isotopes of sulfur or oxygen. However, the large amounts of tracer necessary may not be available and to produce them would require resources greater than those available for this study. For studying transport and dispersion patterns, a conservative tracer is desirable.

Among the potential tracer materials are deuterated methane (CD_4), various perflorocarbons (PFT's) and particulate rare earth oxides. CD_4 and PFT's are conservative tracers; thus conversion of SO_2 to SO_4 and deposition of SO_2 and SO_4 must be accounted for. It has been suggested that non-conservative rare earth particle tracers be used because of their potential to mimic sulfate particles. However, sulfate particles are not directly emitted in significant quantities; rather they are typically formed after considerable transport time which varies with meteorologic conditions. Thus some variable proportion of the rare earth particles will have deposited before the sulfates are formed. Additionally the deposition of SO_2 occurs more rapidly than either sulfate or rare earth particles. Issues such as these must be further investigated before any decision regarding the use of rare earth tracers is made.

CD_4 has low background values and is detectable at very low concentrations, so small amounts of this tracer are sufficient. Though the cost per unit mass is high, the total cost of tracer material is expected to be much less than the cost of PFT's. However, the sample analysis cost is very high ($800-$1000/sample), compared to about $20/sample for PFT's. Thus, it may not be feasible to analyze all samples. If CD_4 were used, samples would be selected for analysis based on air quality and meteorologic data.

The lower analysis costs for PFT's makes it possible to analyze many, if not all of the samples. More information can be obtained regarding the plume position and spatial extent. This would allow a more thorough evaluation of the deterministic modeling. In addition, regression analyses with the receptor models and other statistical analyses would be based on a larger number of samples than if CD_4 were used. With the availability of various PFTs, release times can be staggered such that the age of the samples can be estimated from the sample as well as from trajectory analyses. Alternately, different PFTs could be released from different sources, as previously discussed and the deterministic model results used to estimate plume age.

The SRP tracer study, which used PFT's, apparently had some major problems with the tracer portion of the study. Collocated samplers showed near zero correlation. Four different PFT's were used. The analyses for the first two

PFT's were apparently of better quality than for the third and fourth. The South Coast Air Quality Study (SCAQS) is said to have shown high variability of collocated samples near the detection limits while at the higher concentrations variations of a factor of two were common. There is no theoretical reason that prohibits the use of PFT's or other tracer materials to give quantitative, consistent data. However the pitfalls associated with past experiments demand careful attention and a quality assurance program that monitors the tracer data during the collection process. These issues must be resolved before selection of a tracer approach. A quality assurance plan for tracer release and monitoring, including collection/analysis of background and collocated samples will be developed.

Monitoring

Project MOHAVE field measurements are designed to meet the data requirements discussed in the Data Interpretation and Modeling Section, below. The extinction budget analysis requires data for all of the major particle components (e.g., sulfates, organic and elemental carbon, crustal, and liquid water as estimated from relative humidity) by particle size and concurrent optical parameters (e.g., extinction and scattering coefficient). The attribution analysis requires data for tracer, particle and gaseous sulfur concentrations, particulate trace elements as endemic tracers (e.g., arsenic for smelters and selenium for coal burning), and meteorology (e.g., surface and upper air winds, temperature, and humidity). Additional monitoring of endemic tracers (e.g., methylchloroform for southern California) for non-MPP sources will be conducted to the extent that the resources will allow. Table 1 summarizes the measurements that are anticipated for this program.

To aid in the presentation, monitoring locations have been categorized into several types. Receptor sites are in or near (representative of) GCNP. Monitoring at receptor sites must be capable of supporting extinction budget and attribution analysis. Gradient sites are designed to produce data for attribution analysis. They include sites between sources of interest and GCNP, upwind and background monitoring locations. Upper air meteorological monitoring locations are selected to improve the spatial resolution of the National Weather Service network and to provide vertical wind and temperature profiles in critical areas for input to the deterministic models. Finally aircraft are needed to make tracer and pollutant measurements ranging from near the source to the most distant areas of the study region, and to evaluate vertical distributions.

Table 1. List of the optical variables, aerosol species, meteorological variables and measurement methodologies proposed for the monitoring sites.

Measurement Type	Location	Method	Frequency
Optical			
b_{scat}	A, B, C*	Nephelometer	Continuous
b_{ext}	A	Transmissometer	Continuous
$M_{tf,s}$	A	Photographic	Hourly
Particulate Matter			
Fine Particles			
Mass	A, B, C*	IMPROVE/SCICAS	12 Hours
Ions	A	IMPROVE/SCICAS	12 Hours
Nitrate	A	IMPROVE/SCICAS	12 Hours
Elemental & Organic Carbon	A	IMPROVE/SCICAS	12 Hours
Trace Elements (includes sulfur)	A, B, C*	IMPROVE/SCICAS/SFU	12 Hours
Size Segregating Trace Elements	A	DRUM	12 Hours
Large Particles	A	IMPROVE/SCICAS	12 Hours
Gases			12 Hours
SO_2	A, B, C*	K_2CO_3 Impregnated Filter	12 Hours
Tracer	A, B, C*		12 Hours
Methylchloroform	A, B, C*		
Meteorological - Surface			
Wind Speed & Direction	A, B		Continuous
Temperature, Relative Humidity	A, B		Continuous
Meteorological - Upper Air			
Wind Speed & Direction	Laughlin, Pierce's Ferry	RADAR Profiler	Continuous
Temperature	Laughlin, Pierce's Ferry	RASS	Continuous
Cloud Height and Vertical Pollutant Distribution	Laughlin, Pierce's Ferry	Ceilometer or Upward Looking LIDAR	Continuous

A = Receptor sites, B = Gradient sites, C = Aircraft, * = method and frequency may be different for aircraft

The selection of monitoring locations has an influence on the utility of the data. Table 2 and figure 2 indicate a preliminary list of monitoring locations appropriate for meeting the first objective. An expanded investigation of the impact of all sources of visibility impairment would require additional gradient sites and perhaps additional artificial and endemic tracer measurement capabilities at all sites. Final site selection by the appropriate advisory panel will be influenced by results of simple trajectory analyses run on two years of data (anticipated in March 1991), and practical considerations (i.e., available power, access, security, etc.).

To the maximum degree possible, existing monitoring sites within the study area will be incorporated into the monitoring program. In some cases this would involve providing supplemental equipment or modifying procedures to make data collected at these sites consistent with the other sites in the program. Meteorology data from existing sources (i.e., National Weather Service surface, upper air, and satellite measurements) will be incorporated into the project data base. To the extent that they exist, records of wildfires and prescribed burning and other intermittent source activities will be documented.

Data Interpretation and Modeling

Extinction Budget:

Light extinction is caused by scattering and absorption by particles and gases. In general particle scattering is the principal component of extinction, though in the remote Southwest, scattering by gases that make up the atmosphere (also known as Rayleigh scattering) is a significant fraction on the best air quality days. Black carbon (from diesel engines, forest fires, etc.) is the primary agent of particle absorption, and is occasionally an important cause of haze in the study area. NO_2 is the only common gaseous pollutant that absorbs in the visible portion of the spectrum. It is not expected to play a significant role in Project MOHAVE.

The extinction budget analysis involves determining the contribution to extinction by all of the major contributing components. This can be done statistically using multivariate analysis to explain the optical parameter (b_{ext} or b_{scat}) by a linear combination of the components. These components are the concentrations of the pollutant species multiplied by best-fit determined coefficients interpreted as extinction efficiencies. The hygroscopic particle species (e.g., sulfate and nitrate) include a function of relative humidity to incorporate the effects of water upon the extinction efficiencies of these species. Alternately, first principle calculations (Mie Theory) of the extinction coefficients

Table 2. Possible monitoring sites for intensive and entire study periods by site type.

Site Types	Entire Study	Intensive Only
Receptor	Pierce's Ferry, Meadview*, Hopi Point*, Indian Gardens*, Phantom Ranch*, Long Mesa*	Peach Springs
Gradient	W. Lake Mead, Cottonwood Cove*, Spirit Mtn.*, Overton, Needles, Mojave Desert	Additional sites along Colorado River, southern California, and northern Arizona
Upper Air Meteorology	Laughlin, Pierce's Ferry	Peach Springs, Mojave Desert
Aircraft		Near stack, upwind, along plume, across plume, vertical distribution
* existing NPS and SCE air quality monitoring sites		

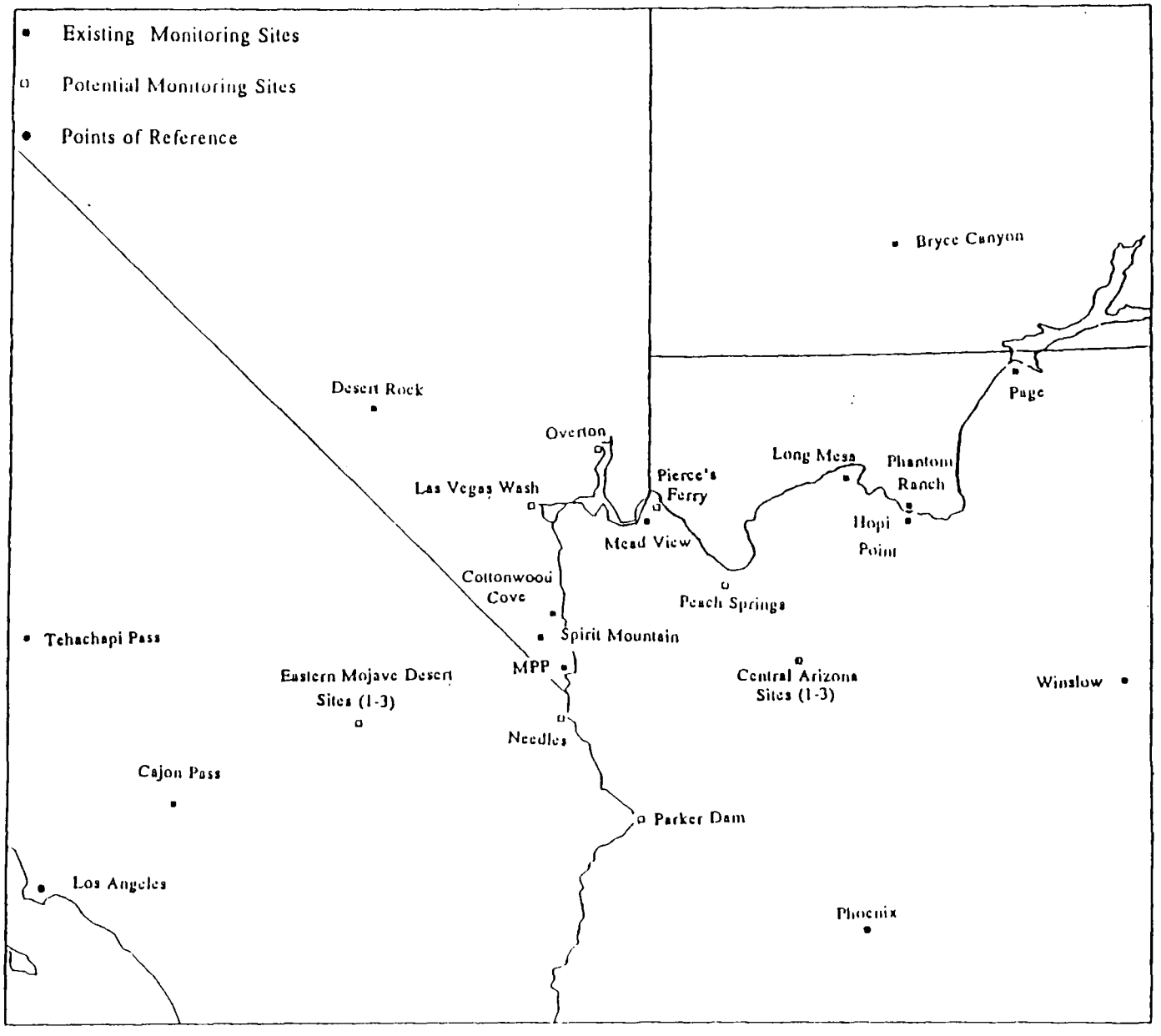


Figure 2. Existing and potential monitoring sites.

can be done, if sufficient particulate characteristics are known (e.g., size distributions). This program will use both procedures and will reconcile the results with literature values of extinction efficiencies.

Attribution analysis:

Attribution analyses will be done using both source-oriented deterministic models and receptor models. Source-oriented deterministic models transport emissions from the source and can account for physical processes en route, including chemical transformation, dispersion and deposition. Receptor models use measurements made at the area of concern (receptors) along with characterization of the emissions from sources potentially affecting the receptor. The contribution of each source to concentrations at the receptors is determined statistically through multivariate analysis techniques which link the sources to the measured concentrations.

Deterministic meteorological and dispersion modeling provides a source-receptor pollution apportionment procedure which is based on fundamental physical conservation relationships. These relationships include conservation equations for velocity, temperature, mass and the three phases of water. The model will provide detailed wind and turbulence fields and a prediction of cloud height and location. Cloud predictions will be checked against satellite photographs and ceilometer measurements, where available.

For Project MOHAVE, deterministic models will be run in an analysis mode using assimilation of the observed data for the entire period of the project. Data assimilation means that measured data will be incorporated into the modeling. The models' utility is to fill in areas between data locations making use of the fundamental physical relationships governing the atmosphere. The incorporation of data assimilation into the deterministic model offers an effective methodology to achieve the best estimate of meteorological transport and dispersion.

The meteorological domain for the simulations will cover the southwestern United States with horizontal grid intervals on the order of 10 km. To obtain better terrain resolution near MPP, a telescoping nested grid will be used. In a nested grid approach, the larger scale results provide the boundary conditions for input into a finer scale modeling domain. In Figure 2, the rectangle bounded by dashed lines "nested" within the larger area demonstrates the concept of a nested grid approach. Horizontal grid intervals in the smallest domain may be

approximately 500 m. For this smaller grid interval, non-hydrostatic models are generally more appropriate than hydrostatic models.

The wind and turbulence fields obtained from the deterministic meteorological model provide the necessary input to calculate the transport and dispersion of the MPP plume. Using this input, a Lagrangian model will be used to transport, disperse and chemically transform the plume. The first step is to transport and disperse the plume; the model results will be compared to the tracer data to evaluate the model. The next step is to incorporate simple chemistry to calculate sulfate concentrations and any other species of interest. The predicted location of emissions from other major sources within the study area will also be identified.

Using complex chemical modeling (e.g. RADM) and explicit inclusion of all the major pollutant sources is very resource intensive and is beyond the scope of objective 1. These analyses may be done as part of objective 2, depending on the level of additional resources.

The use of a deterministic model can also assist in the design of the field program by indicating where instrumentation should be sited and aircraft cross-sections flown so as to optimize the spatial representativeness of the measurements. Also, since the model is based on fundamental concepts, it provides a scientific framework to interpret the data. The same meteorological model simulations can also be used with a wide range of emission inventories in order to assess potential emission control scenarios.

The use of receptor models in apportioning primary particles has been done routinely; however using receptor models to apportion secondary aerosol, as in WHITEX, is more controversial. As in WHITEX, the receptor models used will include the tracer mass balance regression (TMBR) and differential mass balance (DMB) models. These models were designed to estimate the portion of sulfate due to the MPP and the other sources. The results of these receptor modeling will be evaluated in light of the deterministic modeling results. Project MOHAVE will address concerns raised by WHITEX review by making additional measurements and more complete source characterizations. More information on particle size distribution, use of endemic or artificial tracers for other sources, and upper air humidity and cloud height measurements can reduce the uncertainties involved with the use of these models to apportion secondary aerosol. In TMBR and DMB it is assumed that each source has a uniquely emitted tracer associated with it. If not, other methods such as chemical mass balance (CMB) may be first applied to partition the ambient species

concentrations into components attributable to the various groups of sources. TMBR, DMB and CMB are described in detail in the WHITEX report.

Eigenvector analysis, e.g. empirical orthogonal function (EOF) analysis, principal components analysis and factor analysis, will also be done to investigate impacts by specific sources or source areas. The eigenvector analysis results can be used to qualitatively check the deterministic and receptor modeling analyses. Eigenvector analysis shows commonly occurring spatial patterns and their variation in time. The main patterns may be associated with specific sources or source areas. By examining the time series (time variation) of each eigenvector, it can be determined which times a particular source area contributes to concentrations at each site. Meteorologic information, such as wind speed and direction and humidity and its temporal patterns along with source information provides physical information to help interpret and support results of the eigenvector analyses.

The Modeling and Data Analysis technical panel will make specific recommendations concerning the modeling and data analysis approaches to be used.

Extrapolation of intensives study periods to the long-term:

To determine longer term impacts to visibility at GCNP, it is necessary to extrapolate from results of the intensive study periods. This will be a two-step process; the first step will relate the entire 12-15 month study period to the intensive period, while the second will extrapolate from the 12-15 month period to a multi-year period. The first step involves application of source-oriented and receptor models, which are developed and evaluated with intensive period data, to the meteorology and air quality data for the entire study period. In the second step the relative frequency of long-term meteorological patterns will be compared with those during the study period and qualitative adjustments made if necessary.

Source-oriented models will be evaluated and calibrated using the more complete data of the intensive periods. The resulting models will then be run on data from the entire study period. For all modeling analyses a portion of the data may be withheld in order to independently test the models.

During the intensive study periods, receptor modeling will use the artificial tracer results to apportion sulfate due to MPP and any other sources tagged with artificial tracers. Receptor models will also use endemic tracers to apportion remaining significant sources. Results from receptor models based on

endemic tracers will be compared to results of the same models using artificial tracers to evaluate the utility of endemic tracers. If successful, endemic tracer models will then be applied to the entire study period to apportion sources over a complete annual cycle and used in conjunction with the deterministic modeling analysis.

The representativeness of the study year to longer term average conditions will be studied. It should be acknowledged that significant year to year variability in meteorological conditions occurs and that the likelihood of any given year being "typical" is not high. The frequency of occurrence of conditions associated with impacts from each source such as wind speed and direction, humidity, etc. can be compared for the study year and other years for which data are available. Where they exist, optical and air quality measurements from previous years will be compared to the study year measurements. A meteorological classification scheme that uses criteria affecting visibility may be developed. The frequency of occurrence of each pattern for the study period and longer term average can then be compared to put the study year into perspective.

Framework for Summarizing Results:

In a complex program such as this, a sound plan for compilation of results is as important as the collection of high quality and representative data and the performance of appropriate interpretive analysis. Development of an approach to organize the results from this program helps to focus attention and resources on critical steps for the entire program and communicate those ideas to others.

Just as it is inappropriate for worst case results to receive primary attention, it is also inappropriate to dwell on average or typical conditions, especially for an instantaneous effect such as visibility. The 12-15 month study period with hourly deterministic model results requires some method for summarizing the results of the study that avoids these pitfalls. A preliminary conceptual framework for summarizing the results of Project MOHAVE is shown in Table 3. The key idea is the stratification of time periods based upon the locations with respect to GCNP of MPP emissions and those of other significant sources, such as from southern California. These would be based upon the modeling studies. Another stratification is whether the plume(s) has undergone wet or dry chemistry (based upon modeling results and observations). If useful, other stratifications could be developed. The frequency of each condition, the average and standard deviation of the % sulfate from MPP, the % of extinction

Table 3 - Conceptual Framework for Summarizing Project MOHAVE Results

GCNP Impact & Condition	Frequency (Deterministic Model)	% Sulfate (Reconciled Models)	% Extinction (Extinction Budget)	Measure of Perception
No MPP in GCNP				
MPP & SCA Dry				
MPP & SCA Wet				
MPP & Other Sources Dry				
MPP & Other Sources Wet				
MPP Alone Dry				
MPP Alone Wet				
Other Appropriate Categories				

SCA refers to the urban and industrial areas of southern California.

from MPP and a measure of the perceptibility of the MPP impact is estimated for the study period.

Stratification of conditions is expected to not only aid in summarization, but to reduce the uncertainty levels for some of the receptor model results by restricting the variation of parameters assumed to be constant (e.g, chemical conversion rate). Extrapolation to a long-term average may be done through the use of a meteorological classification scheme as previously described. This type of approach provides an efficient manner of presenting the magnitude and frequency of estimated MPP emissions on GCNP over a long-term period that could be used to evaluate the significance of existing impairment.

Appendix 3

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Appendix 4

GROSS APPROXIMATION OF MOHAVE IMPACT TO GRAND CANYON NP VISIBILITY USING HIGHLY SIMPLIFIED ASSUMPTIONS

Assume:

1) Concentration of SO_2 at long distances from source is approximately:

$$SO_2 = \frac{Q}{uhD\tan\theta}$$

Q = SO_2 source strength = 150 tons per day = 1.58 kg s^{-1} = 1.58 X 10^9 ug s^{-1}
u = average wind speed in the mixed layer
h = depth of mixed layer
D = distance from source
θ = lateral plume dispersion in degrees
D tan θ = width of plume at distance D
θ typically 5-15°
distance to GCNP is 120 km
Thus plume width at 120 km = 21 km for θ = 10°, 32 km for θ = 15°
sulfate is $(NH_4)_2SO_4$

2) Determine incremental sulfate concentration that is noticeable:

Assume change in b_{ext} of 10% is noticeable
Scattering efficiency of $(NH_4)_2SO_4$ is 5 m^2g^{-1}

For Rayleigh conditions, b_{ext} = 11 Mm^{-1}, noticeable change is:

$$\frac{1.1\times10^{-6}m^{-1}}{5m^2g^{-1}} = 0.22\mu gm^{-3}$$

For average conditions, b_{ext} = 25 Mm^{-1}, noticeable change is:

$$\frac{2.5\times10^{-6}m^{-1}}{5m^2g^{-1}} = 0.50\mu gm^{-3}$$

CASE 1: prefrontal winter conditions, cloudy

u = 20 m s^{-1} (conservatively high)
h = 3000 m
θ = 10°

$$SO_2 = \frac{1.58\times10^{9}\,\mu g\,m^{-3}}{(2\times10^{1}\,m\,s^{-1})(3\times10^{3}\,m)(2.1\times10^{4}\,m)}$$

$$= 1.25\,\mu g\,m^{-3}\ SO_2$$

$$= 2.6\,\mu m^{-3}\ (NH_4)_2SO_4\ \textit{at 100\% conversion}$$

$$= 1.3\,\mu m^{-3}\ (NH_4)_2SO_4\ \textit{at 50\% conversion}$$

CASE 2: Typical summer afternoon conditions, a) cloudy, b) dry

u = 6 m s^{-1} August average over 18 years at China Lake, 10000 feet MSL
h = 4000 m
θ = 15°

$$SO_2 = \frac{1.58\times10^{9}\,\mu g\,m^{-3}}{(6\,m\,s^{-1})(4\times10^{3}\,m)(3.2\times10^{4}\,m)}$$

$= 2.1\,\mu g\,m^{-3}\ SO_2$ $= 4.2\,\mu g\,m^{-3}$ *sulfate at 100% conversion*

a) cloudy: if $\frac{1}{2}$ SO_2 *contacts cloud, sulfate* $= 2.1\,\mu g\,m^{-3}$

b) dry: assume 3.5% hr^{-1} *conversion*

$$\textit{transport time} = \frac{120\,km}{(6\,m\,s^{-1})\left[3.6\,\frac{km\,hr^{-1}}{m\,s^{-1}}\right]}$$

$= 5.6$ *hours* $= 19\%$ *conversion* $= 0.80\,\mu g\,m^{-3}$

CASE 3: weak pre-frontal winter conditions, cloudy

u = 6 ms^{-1}
h = 1500 m
θ = 15°

$$SO_2 = \frac{1.58 \times 10^9 \mu g m^{-3}}{(6 m s^{-1})(1.5 \times 10^3 m)(3.2 \times 10^4 m)} = 5.5 \mu g m^{-3}$$

$$sulfate = 11.3 \mu g m^{-3} \text{ with } 100\% \text{ conversion } = 5.7 \mu g m^{-3} \text{ with } 50\% \text{ conversion}$$

For the conditions considered, the plume would range from marginally noticeable to quite noticeable using this very simple methodology. The results indicate that further consideration is justified; the potential for impact cannot be dismissed without additional study.

COMPARISON OF AVERAGE SULFUR PARTICULATE GRADIENT BETWEEN SPIRIT MOUNTAIN AND MEADVIEW FOR MPP OUTAGE AND NON-OUTAGE CONDITIONS

The difference between average particulate sulfur concentrations at Spirit Mountain and Meadview were compared for outage and non-outage conditions. The gradients were compared for all wind directions and for wind directions transporting the plume toward the site. Data is from Murray, et al, 1989.

All wind directions

Site	MPP status	S μgm^{-3}	n
Meadview	off	0.363	36
	on	0.383	454
Spirit Mtn	off	0.410	54
	on	0.385	442

Wind direction $\leq \pm 90°$ from MPP to site

Site	MPP status	S μgm^{-3}	n
Meadview	off	0.363	36
	on	0.379	358
Spirit Mtn	off	0.508	25
	on	0.439	242

For all wind directions : $Spirit_{on} - Meadview_{on} = 0.002\ \mu gm^{-3}$
$Spirit_{off} - Meadview_{off} = 0.047\ \mu gm^{-3}$

Average difference in gradient= $0.045\ \mu gm^{-3}$

For wind direction $\leq \pm 90°$: $Spirit_{on} - Meadview_{on} = 0.060\ \mu gm^{-3}$
$Spirit_{off} - Meadview_{off} = 0.145\ \mu gm^{-3}$

Average difference in gradient= $0.085\ \mu gm^{-3}$

It can be seen that the average gradient in particulate sulfur between Spirit Mountain and Meadview is greater when MPP is not operating, particularly for wind directions favorable for transport from MPP. It is hypothesized that the gradient is small when MPP is operating because increased dilution of the southern California sulfur between Spirit Mountain and Meadview is balanced by formation of particulate sulfur in the MPP plume. During outage

conditions, this does not happen; thus the gradient between Spirit Mountain and Meadview is increased.

The difference in gradient of 0.085 μgm^{-3} particulate sulfur corresponds to about 0.34 μgm^{-3} sulfate as ammonium bisulfate. Assuming a mass scattering efficiency of 5 m^2g^{-1}, this would add an average of 1.7 Mm^{-1} to the extinction coefficient. This increase would be expected to be marginally perceptible for very clear days ($b_{sp} \leq 17\ Mm^{-1}$) and imperceptible for other days. However, these estimates are for concentrations averaged many days, while visibility is likely to vary significantly over the course of a day, and between days.

It should be emphasized that the data base used is very limited and no conclusions can be made regarding the impact of MPP using this limited data set. However, the difference in gradients when MPP is off compared to on suggests the hypothesis put forth above may be correct.

August 26, 1991

MOHAVE OUTAGE STUDY--A PLAN SYNOPSIS

Prepard by:

Desert and Intermountain Air Transport Program

The appropriation by Congress of 2.5 million dollars to EPA to conduct a source apportionment study on the Mohave Power Project (MPP) has generated widespread interest in the analysis of the data obtained during a seven month period the MPP was inoperative in 1985. This outage period represents the ultimate in experiments. The plant was turned off and the effects can be examined. It serves as a baseline for assessing the impact of the MPP on visibility degradation in the Grand Canyon National Park (GCNP). The data base can also be used in other statistical analyses that utilize the fluctuating power plant load as a sort parameter.

The initial study was published by Murray et al. (1989) which showed that the sulfate concentration at Meadview was not significantly lower than that observed during similar periods in other years. The study set an upper bound to the sulfate due to MPP observed at Meadview as less than 15%. The power of this analysis was limited by its rudimentary treatment of inter-annual meteorological differences. The paper's impact is limited by the fact that the authors did not consider the daily power plant load during the control periods.

The SCENES data base was utilized in the analysis of the outage period with respect to similar periods in other years. That program was designed and implemented to acquire high quality data for studies of visibility degradation.

The outage study used the 24-hour average particulate samples at three sites, one background and two receptor, with respect to impact from MPP. Chemical and physical analyses of the filters were carried out only on every third day. The samples for the two intermediate days were archived.

A re-examination of the outage and other periods of reduced power plant output compared to periods when the plant operates at or near capacity during the SCENES program is envisioned. The new study will incorporate the data used in the original analysis but would also embody the following elements:

- Independent statistical analysis of the experiment.

- Chemical analysis of all the filters. (Quality assurance will be evaluated through comparison of current results to past data through regression and time series analysis.)

- Classification of the synoptic weather patterns affecting transport from MPP to GCNP.

- Deterministic modelling of the wind flow patterns associated with each of the meteorological regimes.

- A detailed compilation of regional SO_2 emissions data for the control and outage periods. (Changes in emission patterns must be included in the final analysis.)

These elements will be described in the following sections.

Statistical Analyses--Dr. Paul Switzer of Stanford University will serve as the independent statistician. He has a history of involvement in physical measurement processes. He will:

- Be responsible for the overall experiment design after consultation with the principal scientists (Hoffer, White and Koracin), the participants in the original study and familiarization with the existing data base. (The written experiment design document would be a cooperative effort.)

- Be responsible for the specification of the techniques used to handle the data and the statistical tests that will be applied. All data manipulations can be performed in the "blind". This procedure has been used in the past within the meteorological community to evaluate the results of weather modification experiments and has proven effective in eliminating cries of bias and data selection.

- Be responsible for sample handling procedures (if the samples are assigned random numbers), data stratification, application of statistical tests and reporting of the results.

- Participate in the redesign of the meteorological classification scheme, assisting with the number of synoptic categories needed for stratification and in defining the variability limits within categories when the wind field is applied in the deterministic modelling effort. The statistician in consultation with the principal scientists, will set the limits on the meteorological data stratification.

- Participate with the individuals who have contributed substantially to the project in the preparation and submission of a research paper to a peer reviewed journal.

Chemical Analysis--All the filters including those already analyzed will be analyzed using XRF. The contractor will perform the analysis and report the information after the sample date has been replaced by a random number supplied by the statistician or his agent. Sample random numbers would be attached to the filters by the following procedure:

- A list of dates versus random numbers would be prepared by the statistician.

- The sample ID numbers corresponding to the dates would be used to generate a list of sample ID versus random number. The sample ID would be replaced by the random number using the following procedure:

  - Two individuals not associated with the project would travel to Oregon (NEA) to handle the samples within the contractor's facility.

  - The first individual would place the random number associated with the ID number on the sample container.

  - The second individual would check to be certain that the two numbers were correct before removing the ID number.

The primary element of interest to this study is sulfur. Selenium, arsenic and the other trace elements are of secondary importance. These elements are stable, so the quality of the sample should not have degraded with time. The filter analyses will be performed through the external contractor who performed the original analysis.

Sulfur The sulfate concentration will be determined by measuring elemental sulfur using XRF at an intermediate protocol, Protocol 5.

Arsenic, Selenium and Other Trace Elements The trace element concentrations will be obtained from the XRF data. If at the end of the experiment it becomes essential to use additional elements as tracers, arsenic and selenium could be determined using neutron activation. Arsenic has a short half-life and will be counted by the contractors. The long half-life of selenium as well as other long half-life elements will be counted at DRI to lower the overall costs of sample analysis.

Meteorological Classification--The sampling period was a 24-hr day, from midnight to midnight, starting in June 1985. Some adjustments in data handling will be made for the data taken on 8 and 16 hour increments prior to 1985. The meteorological conditions prevailing during each sampling period will be classified using the meteorological classification scheme developed by Farber et al. (1989) with some modification to incorporate more surface data and a probability of the occurrence of cloud, based upon surface observations and upper air observations. All sample days will be included in the computer calculations of the classification probabilities. As a part of the classification, a parameter quantifying the strength of the synoptic flow, such as geostrophic wind, height gradient or vorticity, will be tabulated. Statistical analysis of the strength parameter will be used to define limits on the wind speed and direction parameters used in deterministic modelling.

Deterministic Modelling--A minimum of two meteorological models with the appropriate grid spacing (telescoping grid starting at 1 km) will be exercised for each of the meteorological classifications. The strength of the synoptic flow determined from the classification analysis will serve as an input to the model. The wind speed and direction parameters and their variance will be fixed prior to running the models. At the present

time, the addition of a chemical module to the meteorological model is not contemplated. However, should a good chemical module become available it would be exercised along with the meteorological model.

The transport modelling will be used to assign nominal MPP impacts at Meadview. A potential dosage (concentration x time) will be calculated from the simulated dispersion and duration of the plume at Meadview. Calculations will be performed for all classifications and synoptic strengths, yielding a nominal MPP impact corresponding to each sampling interval. These daily nominal impacts will serve as input variables to the statistical analysis, inputs that incorporate all relevant meteorological information in a physically correct way.

SO_2 Emissions--A subcontract will be awarded to an outside contractor specializing in emissions inventory following a competitive solicitation. The firm will inventory the regional SO_2 emissions and report the results by month and subregion. The inventory will be compiled for all types of sources for the period of the study, and will be used as a guide to regional changes in the background SO_2/SO_4 concentrations.

Project Personnel--The project would be undertaken by the Desert and Intermountain Air Transport Program (DMAT) under the sponsorship of Southern California Edison Company (SCE). The project manager will be Dr. Thomas Hoffer, the coordinating scientist Dr. Warren White, the statistician Dr. Paul Switzer and the deterministic modellers Drs. Leif Enger, Darko Koracin and David Rogers. The synoptic classification will be undertaken by a team comprised of Dr. David Rogers, Dr. Mark Green, Dr. Rob Farber and Sara Pryor.

Summary--A reanalysis of data collected during the MPP outage is proposed to refine and strengthen the bound on MPP's contribution to haze in the GCNP. The experiment, as proposed, will strive to eliminate bias in the application of data stratification and statistical analysis.

The project schedule calls for an immediate start with a spring 1992 completion date.

Appendix 6

THE CSU RAMS

INTRODUCTION

The numerical atmospheric models developed independently under the direction of William R. Cotton and Roger A. Pielke have recently been combined into the CSU Regional Atmospheric Modelling System (RAMS). Development of many of the physical modules has been accomplished over the past 15 years and has involved over 50 man years of effort. RAMS is a general and flexible modelling system rather than a single purpose model. For example, current research using RAMS includes atmospheric scales ranging from large eddy simulations ($\Delta x \approx 100\ m$) to mesoscale simulations of convective systems ($\Delta x \approx 100\ km$). This paper will discuss the options available in RAMS, the engineering aspects of the system and how the flexibility is attained.

RAMS OPTIONS

RAMS is a merging of basically three models that were designed to simulate different atmospheric circulations. These were a non-hydrostatic cloud model (Tripoli and Cotton, 1982) and two hydrostatic mesoscale models (Tremback *et al.*, 1985 and Mahrer and Pielke, 1977). The capability of RAMS was recently augmented with the implementation of 2-way interactive grid nesting. Because of this, the modelling system contains many options for various physical and numerical processes. These options are listed below.

The following options are currently available in configuring a model:

1. Basic equations:

   Option 1 Non-hydrostatic time-split compressible (Tripoli and Cotton, 1980)

   Option 2 Hydrostatic incompressible or compressible (Tremback *et al.*, 1985)

2. Dimensionality: 1, 2, or 3 spatial dimensions

3. Vertical coordinate:

   Option 1 Standard cartesian

   Option 2 Sigma-z

4. Horizontal coordinate:

   Option 1 Standard cartesian

   Option 2 Polar stereographic

5. Grid Structure:

   - Arakawa-C grid stagger
   - Unlimited number of nested grids
   - Unlimited number of levels of nesting
   - Ability to add and subtract nests
   - Moveable nests

6. Finite differencing:

   Option 1 leapfrog on long timestep, forward-backward on small timestep, 2nd or 4th order flux conservative advection.

   Option 2 forward-backward time split, 2nd or 6th order flux conservative advection (Tremback *et al.*, 1987)

7. Turbulence closure:

   Option 1 Smagorinsky-type eddy viscosity with R_i dependence

   Option 2 Level 2.5 type closure using eddy viscosity as a function of a prognostic turbulent kinetic energy

   Option 3 O'Brien profile function in a convective boundary layer (Mahrer and Pielke, 1977); local exchange coefficient in a stable boundary layer (McNider, 1981).

8. Condensation

   Option 1 Grid points fully saturated or unsaturated

   Option 2 No condensation

9. Cloud microphysics

   Option 1 Warm rain conversion and accretion of cloud water (r_c) to raindrops (r_r), evaporation and sedimentation (Tripoli and Cotton, 1980)

   Option 2 Option 1 plus specified nucleation of ice crystals (r_i), conversion nucleation and accretion of graupel (r_g), growth of ice crystals (r_i), evaporation, melting and sedimentation (see Cotton *et al.*, 1982)

   Option 3 Option 1 plus option 2 plus predicted nucleation and sink of crystal concentration (N_i), conversion and growth of aggregates (r_a), melting, evaporation and sedimentation. The nucleation model includes: sorption/deposition, contact nucleation by Brownian collision plus thermophoresis plus diffusiophoresis, secondary ice crystal production by rime-splinter mechanism (Cotton *et al.*, 1986).

   Option 4 No precipitation processes

10. Radiation:

    Option 1 Shortwave radiation model including molecular scattering, absorption of clear air (Yamamoto, 1962), ozone absorption (Lacis and Hansen, 1974) and reflectance, transmittance and absorptance of a cloud layer (Stephens, 1978), clear-cloudy mixed layer approach (Stephens, 1977). (See Chen and Cotton 1983, 1987.)

    Option 2 Shortwave radiation model described by Mahrer and Pielke (1977) which includes the effects of forward Rayleigh scattering (Atwater and Brown, 1974), absorption by water vapor (McDonald, 1960), and terrain slope (Kondrat'yev, 1969).

    Option 3 Longwave radiation model including emissivity of a clear atmosphere (Rodgers, 1967), emissivity of cloud layer (Stephens, 1978), and emissivity of "clear and cloudy" mixed layer (Herman and Goody, 1976)

Option 4 Longwave radiation model described by Mahrer and Pielke (1977) including emissivities of water vapor (Jacobs *et al.*, 1974) and carbon dioxide (Kondrat'yev, 1969) and the computationally efficient technique of Sasamori (1972).

Option 5 No radiation

11. Transport and diffusion modules:

    Option 1 Semi-stochastic particle model for point and line sources of pollution (McNider, 1981)

12. Lower boundary:

    Option 1 Surface layer similarity theory based on Louis (1979) as a function of specified surface roughness over land and predicted sea surface roughness based on Garratt and Brost (1981).

    Option 2 Surface layer temperature and moisture fluxes are diagnosed as a function of the ground surface temperature derived from a surface energy balance (Mahrer and Pielke, 1977). The energy balance includes longwave and shortwave radiative fluxes, latent and sensible heat fluxes, and conduction from below the surface. To include the latter effect, a multi-level prognostic soil temperature model is computed.

    Option 3 Modified form of Option 2 with prognostic surface equations (Tremback and Kessler, 1985)

    Option 4 Same as Option 2, except vegetation parameterizations are included (McCumber and Pielke, 1981; McCumber, 1980)

13. Upper boundary conditions:

    Option 1 Rigid lid (non-hydrostatic only)

    Option 2 Rayleigh Friction layer plus Option 1-4

    Option 3 Prognostic surface pressure (hydrostatic only)

    Option 4 Material surface top. (hydrostatic only) (Mahrer and Pielke, 1977)

    Option 5 Gravity wave radiation condition (Klemp and Durran, 1983)

14. Lateral boundary conditions:

    Option 1 Klemp and Wilhelmson (1978a,b) radiative boundary conditions

    Option 2 Orlanski (1976) radiative boundary conditions

    Option 3 Klemp and Lilly (1978) radiative boundary condition

    Option 4 Option 1, 2 or 3 coupled with Mesoscale Compensation Region (MCR) described by Tripoli and Cotton (1982) with fixed conditions at MCR boundary

    Option 5 The sponge boundary condition of Perkey and Kreitzberg (1976) when large scale data is available from objectively analyzed data fields or a larger scale model run. This condition includes a viscous region and the introduction of the large scale fields into the model computations near the lateral boundaries.

15. Initialization

Option 1 Horizontally homogeneous.

Option 2 Option 1 plus variations to force cloud initiation.

Option 3 NMC data and/or soundings objectively analyzed on isentropic surface and interpolated to the model grid.

Option 4 NMC data interpolated to the model grid.

As one can see, RAMS is quite a versatile modelling system. RAMS has been applied to the simulation of the following weather phenomena.

1. Towering cumuli and their modification
2. Mature tropical and mid-latitude cumulonimbi
3. Dry mountain slope and valley circulations
4. Orographic cloud formation
5. Marine stratocumulus clouds
6. Sea breeze circulations
7. Mountain wave flow
8. Large eddy simulation of power plant plume dispersal
9. Large eddy simulation of convective boundary layer
10. Urban circulations
11. Lake effect storms
12. Tropical and mid-latitude convective systems

ENGINEERING ASPECTS

Because of the large number of options in RAMS, the structuring of the code needs to be carefully considered. This section will discuss various aspects of the code structure of the system.

Pre-processor The code of RAMS is written in as close to the FORTRAN 77 standard as possible. However, with a program as large as this, the FORTRAN standard is lacking in several features such as global PARAMETER and COMMON statements and conditional compilation. To remedy these insufficiencies, the RAMS code takes advantage of a pre-processor written as part of the RAMS package. This pre-processor itself is written in the 77 standard so that the package as a whole is highly portable. It takes full advantage of the character features of FORTRAN and has executed successfully on a number of machines including a VAX, CRAY-1, CRAY-X-MP, and CYBER 205 without modification. Some of the features of the pre-processor are described below:

1) By including a character in the first column of a line of code, that line can be "activated" or "eliminated" from the compile file. This allows for conditional compilation of single lines or entire sections of code.

2) A pre-processor variable can be set to a value. This variable can then be used in other expressions including a pre-processor IF or block IF to conditionally set other pre-processor variables. These variables also can be converted to FORTRAN PARAMETER statements which can be inserted anywhere in the rest of the code.

3) A group of statements can be delineated as a "global" which then can be inserted anywhere in the code. This is very useful for groups of COMMON and PARAMETER statements.

4) DO loops can be constructed in a DO/ENDDO syntax, eliminating the need for statement labels on the DO loops.

Two-way interactive grid nesting The use of grid nesting allows a wider range of motion scales to be modeled simultaneously and interactively. It can greatly ease the limitations of unnested simulations in which a compromise must be reached between covering an adequately large spatial domain and obtaining sufficient resolution of a particular local phenomenon. With nesting, RAMS can now feasibly model mesoscale circulations in a large domain where low resolution is adequate, and at the same time resolve the large eddy structure within a cumulus cloud in a subdomain of the simulation.

Nesting in RAMS is set up such that the same model code for each physical process such as advection is used for each grid. This makes it easy for any desired number of grids to be used without having to duplicate code for each one. Also, it is easy to add or remove a nested grid in time, and to change its size or location. There is still the flexibility of choosing many model options independently for different grids.

RAMS has adopted the two-way interactive nesting procedure described in Clark and Farley (1984). This algorithm is the means by which the different nested grids communicate with each other. The process of advancing coarse grid A and fine nested grid B forward in time one step begins with advancing grid A alone as if it contained no nest within. The computed fields from A are then interpolated tri-quadratically to the boundary points of B. The interior of B is then updated under the influence of its interpolated boundary values. Finally, the field values of A in the region where B exists are replaced by local averages from the fields of B. An increase in efficiency over the Clark and Farley method was implemented by allowing a coarse grid to be run at a longer timestep than a fine grid.

The following options are available with nesting in RAMS:

1) There is no imposed limit (only a practical one) to the number of nested grids which can be used.

2) When two grids B and C are nested within grid A, they may be either independent (occupying different space) or C may be nested within B.

3) The increase in spatial resolution of a nested grid may be any integer multiple of its parent grid resolution. Moreover, this multiple may be specified independently for the three coordinate directions.

4) A nested grid may, but need not, start from the ground and extend to the model domain top.

5) A nested grid may be added or removed at any time during a simulation.

6) A nested grid can travel horizontally at a prescribed velocity.

I/O structure For those machines with limited central memory and a "non-virtual" operating system or for efficiency on virtual systems, RAMS is constructed with a disk I/O scheme. When the scheme is operating, a subset of the model's three-dimensional variables will reside in central memory at any one time. Computations then can be performed with this subset. When these computations are finished, a new subset of three-dimensional variables are requested and computations performed with these. The RAMS structure, thus, is dependent on this I/O scheme and consists of a series of calls to the I/O scheme and to the routines which do the calculations.

Modularity For flexibility, RAMS is written as modular as possible. Each individual physical parameterization or numerical process is put in a separate subroutine so that the routines can easily be replaced for different options or with new developments.

Computational routines The routines that do the actual computations for the model are written so that the implementor of a new or replacement routine does not need to be concerned with most of the details of the rest of the model computations. All three-dimensional variables are "passed" to the subroutines through the call statement with other variables passed through COMMON. The implementor then has the flexibility to structure his routine in whatever manner he wishes to produce the desired result. This concept will also make the implementation of routines from other models and programs easier with less modification required.

Analysis routines A set of subroutines has been developed for analyzing and plotting a variety of quantities from fields output from RAMS. This greatly facilitates the interpretation and understanding of modeled atmospheric phenomena. The quantities diagnosed by these routines include vorticity, divergence, streamfunction, energy, momentum flux, most variances and covariances, and layer averaged quantities.

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1. DEVELOPMENT OF AN INTERACTIVE DATA ANALYSIS TOOL USING THE MONTE CARLO MODEL

1.1 INTRODUCTION

The 1990 Clear Air Act explicitly recognizes the existence of long range transport of air pollution. Several provisions of this significant new law require regulatory actions that involve multi-state regions, dictated by regional-scale air pollution. For instance, the Act requires the establishment of Transport Commissions over the next five years. These commissions will be charged with the policy developments for "airshed" on regional scale involving several neighboring states.

The work of such commissions and many other provisions of the 1990 law has a strong need for technical input on the nature and scope of regional air pollution. Typical questions may be: What is the region of influence for specific sources; What are the major source regions contributing to a given receptor; How will certain emission reduction scenarios reflect on ambient pollution levels.

In the past, the answers to such questions have been obtained either form intensive monitoring and measurement campaigns or from prognostic regional models. Intensive measurement programs are expensive and generally provide answers applicable only to measurement domain. Prognostic models, on the other hand, are in general rather unreliable. Hence, for the effective implementation of the new law, new approaches and tools are needed. The PC diagnostic Monte Carlo model proposed for this project, will provide such a policy oriented data analysis/interpretation.

Over the past two decades, much scientific knowledge has been gathered about the nature and scope of regional air pollution. In fact, it can be stated that the main causes, and physico-chemical processes that characterize regional air pollution are reasonably well understood.

The CAPITA diagnostic Monte Carlo model, encapsulates and describes much of the knowledge about regional sulfur pattern. It was developed in the early 1980s for the analysis and interpretation of regional sulfur and visibility data. It's application to other areas are illustrated in section 1.6.

The initial version of the CAPITA model served primarily as a research tool. In the 1990s there will be a strong need for operational, easy to use well calibrated models that can aid the implementation of the complex new Act. The proposed PC Monte Carlo model is intended to be a tool to aid policy-related decision making.

1.2 PURPOSE

The purpose of this task is to develop an interactive and physics-based data analysis tool for the analysis and interpretation of visibility related data. The data analysis tool is to aid policy oriented and scientific decision making. The results of the work should be directly applicable to the implementation of the 1990 Clean Air Act.

1.3 GOALS

The project has the following specific goals:

a. Implement a personal computer-based version of the CAPITA Monte Carlo regional model (PCMC)
b. Re-examine the calibration of the diagnostic model using more recent high quality aerosol, gaseous, precipitation chemistry, and visibility related data sets.
c. Develop interfaces to meteorological transport data produced by other, more elaborate meteorological models.
d. Present the results suitable for answering policy questions, such as those posed by the new Clean Air Transport Commissions.
e. Develop interactive graphic user interface that will:
   aid the operation of the model by non-programmers
   facilitate the graphic display of results
   - presentation of the physical entities, such as spatial concentration maps, time charts, frequency distributions
f. As much as possible, use off-the-shelf robust software building blocks in the creation of the interactive PC Monte Carlo model.

1.4 SCOPE

The scope of this task will include the porting, testing, and re-calibration of the CAPITA Monte Carlo model on a PC platform. It also involves building user interfaces for the input/output of model data.

In this task, the main scientific/regulatory application of the PCMC model will be to visibility. This work will not involve significant new research areas in of atmospheric processes, policy analysis or other fields. Rather, it will use the available knowledge in these areas and package these into generally usable PC tools.

1.5 EXPECTED RESULTS

The results of this task will be an interactive data analysis and presentation package that will allow the simulation modeling of visibility-related atmospheric processes. The model will be packaged as a tool. As a tool it should be usable by policy analysts

within and outside the government as well as by the research community.

1.6 THE CAPITA MONTE CARLO MODEL

The proposed interactive data analysis tool will utilize the CAPITA Monte Carlo regional atmospheric transport/transformation/removal model. The model principles and some of its applications are described by Patterson et al. (1981). The following description states the concept of the model and illustrates some of its past applications relevant to this work.

In the Mote Carlo modelling approach, simulated pollutant quanta (particles) are "emitted" in accordance with an emission inventory. These quanta are moved in fixed time increments using the interpolated measured wind fields. During their transport, the pollutant quanta may be subject to chemical transformations or removal. The transport as well as the transformations are somewhat randomized, hence the name Monte Carlo method. The method is also referred to as the *Direct Simulation* method since the physico-chemical processes are simulated as discrete events rather than obtained from the solution of differential equations. The result of Monte Carlo simulations is a large number (10^5 - 10^6) of pollutant "particles" dispersed geographically for every time step of the simulation.

Table 1. Summary of CAPITA regional model

a. Model Type	Monte Carlo (Lagrangian, and Eulerian in the limit of large number of quanta)
b. Receptor Grid	52 x 60 grids (variable for PCMC)
Grid Resolution	127 x 127 km at 60 degrees north latitude (one-third of U.S. National Meteorological Center grid spacing)
c. Model Domain	North America (variable for PCMC)
d. Model Output	Fields of particles each 3-h, each particle representing a mass of emitted pollutant remaining in the atmosphere as each possible chemical species; converted to fields of daily SO_2, SO_4 concentration and dry and wet depositions at all grid points
e. Input requirements	
Emissions	Seasonal, surface and tall stack 1890-~~1980~~ 2010 SO_2 grids.
Winds	0000 and 1200 GMT rawinsonde wind profiles at 130 sites. In PCMC, externally generated wind fields accepted
Precipitation	3-hourly observation of precipitation of three intensities at surface synoptic sites.
Cloud cover	gridded from 3h surface synoptic sites
Dewpoint	gridded from 3h surface synoptic sites
Mix heights	Climatological by season, from work of Holzworth (1972) and Portelli (1977), of maximum afternoon mixing heights
f. Emissions	3-h SO_2 emissions, Released in mixed layer in day, and either 150-450 layer or 0-150m at night; 1% primary SO_4
g. Transport	Inverse-distance squared weighting. Upper air rawinsonde winds are interpolated in space into 11 layers (0-150, 150-450, 450-750, 750-1050, 1050-1350, 1350-1650, 1650-1950, 1950-2250, 2250-2850, 2850-3450, 3450-5250 m above ground). Wind for each layer is the vector average, and winds are linearly interpolated from 12h to 3h, using seasonal diurnal interpolation factors at each height to reflect nighttime jets and midday drag from convective mixing.
h. Precipitation	3h grids of the space-time average probability of encountering precipitation are used to scale local wet removal rates as fraction of the maximum rate.
i. Mixed Layer	Climatological average, which varies with geographic location and season of year. Representative peak P.M. values are 800 (winter), 1200 (spring, fall), 1350 (summer). Fixed 150m at night.
j. Horizontal Dispersion	Lateral displacement by veering of layers overnight; "eddy diffusion" K=2000 m^2/s day, 100 m^2/s night.
k. Vertical Dispersion	instantaneously mixed throughout mixed layer during day (0900-1800 LST); no vertical mixing at night.
l. SO_2 Transformation Rate	Varies seasonally, diurnal and locally. "Dry" part proportional to solar radiation, function of latitude/season, time of day, and local total sky cover. "Wet" part proportional to local surface dewpoint.
m. Dry Deposition Rate	Zero above local mixed height and above 150m night surface layer. Varies with stomatal density and opening.
n. Wet Removal Rate	Zero above local mixed height within mixing layer; (precipitation probability over a grid during a specified time) x (wet removal rate constant, 100%/h for SO_4 and 10%/h for SO_2).

In the PC implementation, some of the above model parameters will be changed. The changes will incorporate better physico-chemical knowledge, better computational performance and a more general user interface. In what follows, the application of the Monte Carlo model in different domains is illustrated. We consider that the illustrations below demonstrate the potential applications of the new PC based model.

Receptor Modeling and Back Trajectory Analysis. The simplest application of the model is for showing back trajectories leading to a specific receptor site. The approach is illustrated below as applied to the analysis and interpretation of the measurements in the VISTTA program (Macias et al. 1981).

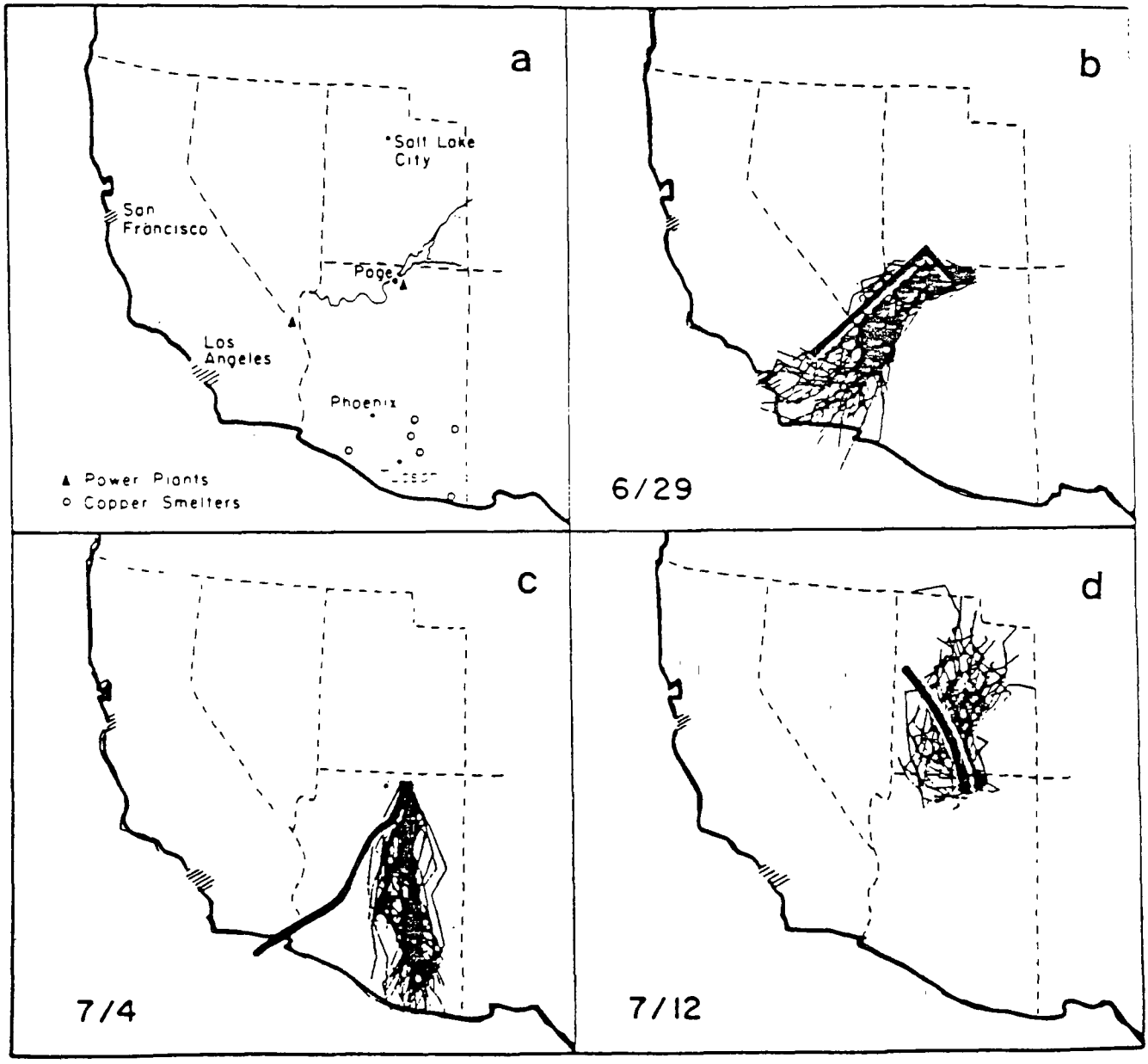


Fig. 1. (a) Map of the portion of the southwestern U.S. of interest in this study. Some of the major emission sources are indicated. (b) (c) and (d) Intercomparison of calculated air mass histories. Each figure shows two estimates for the history of air sampled at Page at 11.00 MST on the indicated date. Single heavy tracks are back trajectories derived by meteorologists from measurements of upper-air winds (MRI, 1980). Multiple light tracks are back trajectories computed by CAPITA Monte Carlo model from adjusted midday surface winds, taking dispersion into account (Patterson *et al.*, 1980). The two estimates, based on independent manipulation of independent data sets, agree satisfactorily for the three differing transport regimes shown here, and for 16 of the 18 days considered.

Multiple Plume Modeling. The model was also applied for the modeling of single and multiple plume dispersion. The figure below indicates the model usage for the visualization of multiple plumes in the Southwestern U.S. (Macias et al., 1981).

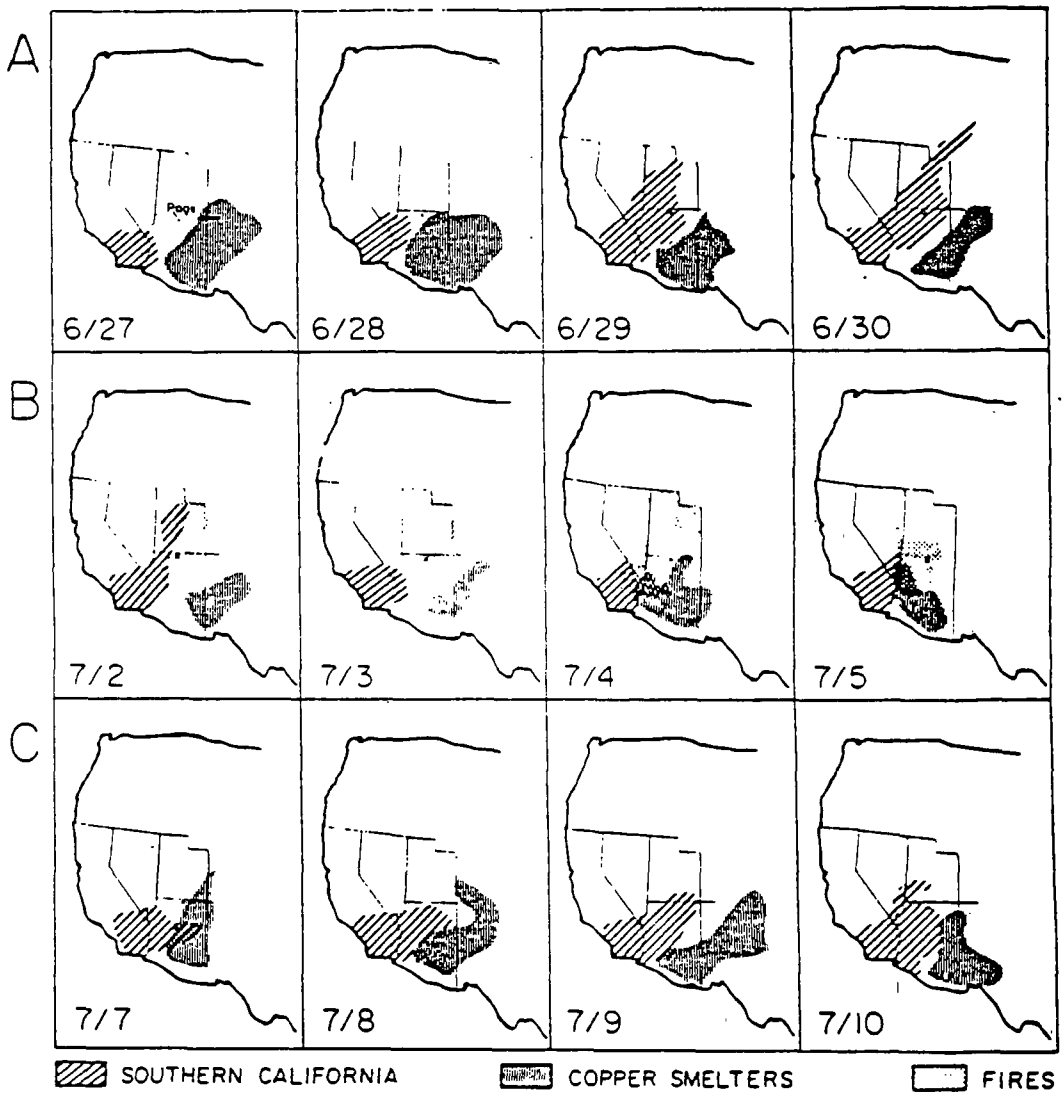


Fig. 2. Calculated plumes from potential source areas, from CAPITA Monte Carlo model. Each figure shows approximate extent, at 11.00 MST on the indicated date, of material released during the preceding 48 h (A, C) or 24 h (B). The three rows cover the three characteristic time periods identified by Macias *et al.* (1981). (A) 27–30 June: Air which had stagnated over southern California for several days moved into the Page area during the latter half of this period. (B) 3–6 July: Shifting southerly winds brought material to the vicinity of Page from wildfires north of Phoenix and smelters southeast of Tucson. (C) 8–11 July: A shift to more southerly winds during the latter half of this period diminished the impact of southern California on Page.

Regional Transmission Modeling. The most extensive use of the CAPITA model has been the regional modeling of sulfates and extinction coefficients over the eastern U.S. (Patterson et al., 1981). In that application, the daily pattern for sulfate aerosol and extinction coefficients were simulated as shown in the figure below.

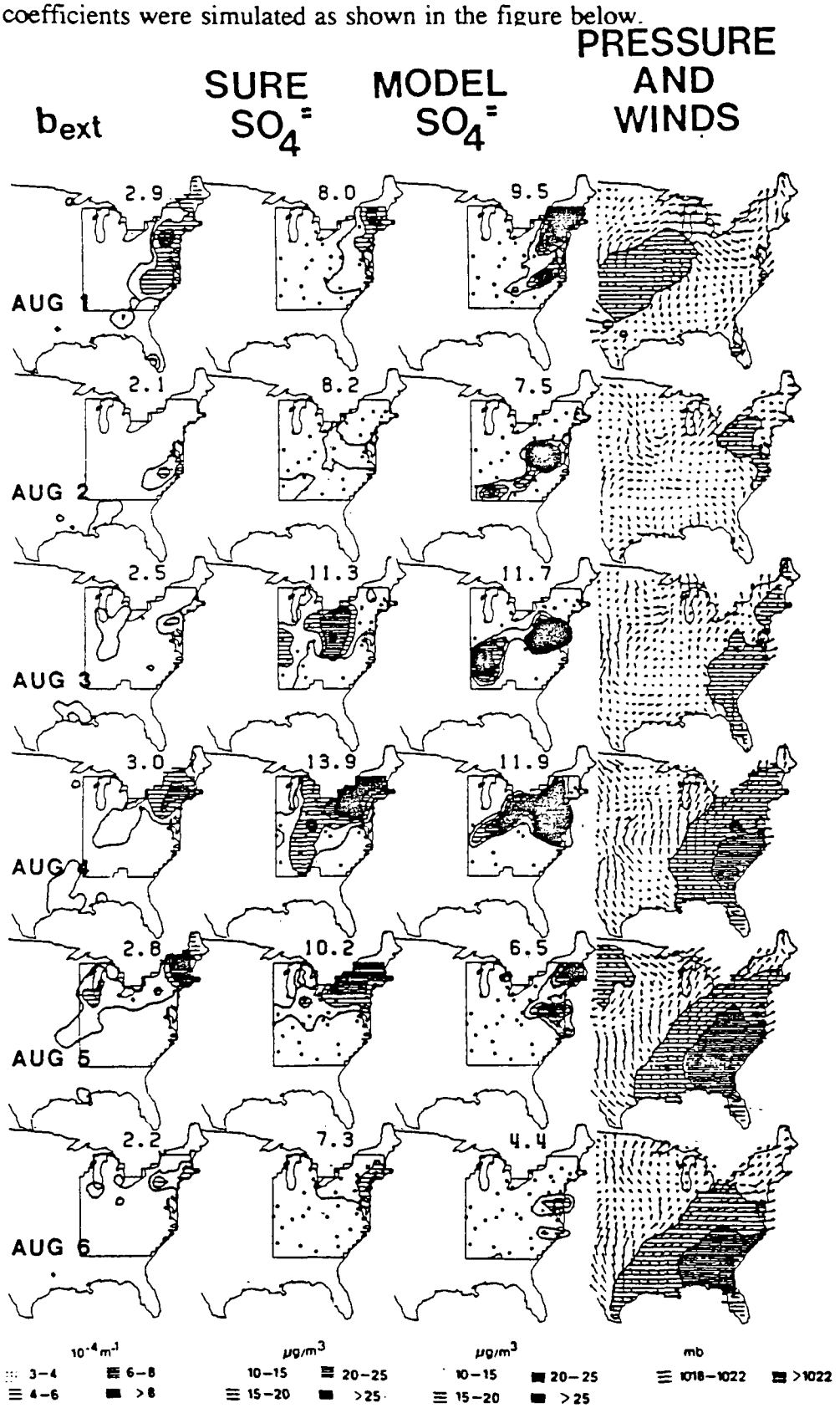


FIG. 3. Daily maps of midday b_{ext} corrected to 60% RH (first column), 24 h average SURE $SO_4^=$ (2nd column), modeled 24 h distribution of emitted sulfur quanta (third column), and unmodified noon surface wind field overlaid with the sea level pressure (last column) for 1–6 August 1977.

The daily pattern of measured SO_4, model SO_4, visual range-derived extinction coefficient, b_{ext}, and air residence time is shown in Figure 4.

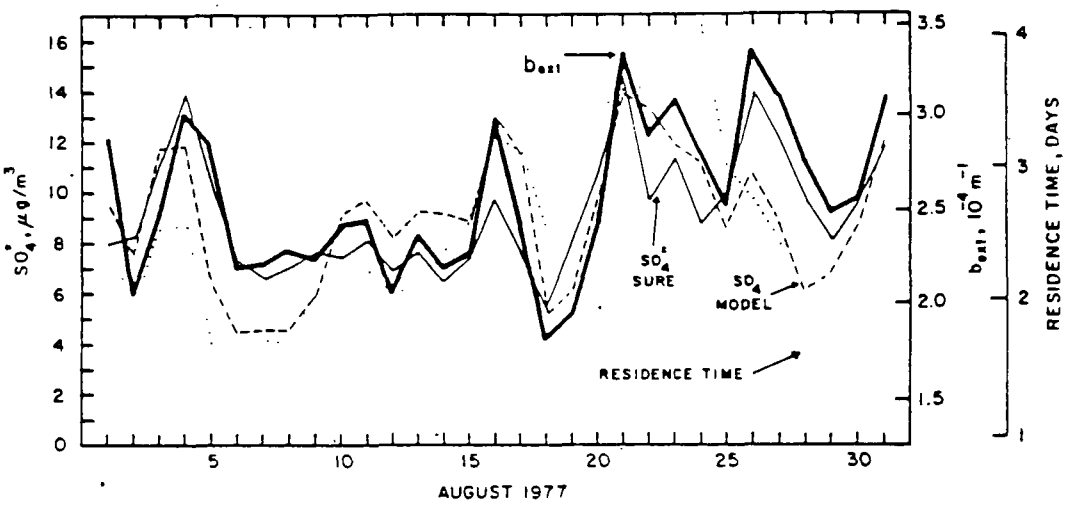


FIG. 4. Daily spatial averages within the SURE region of SURE sulfate (thin line), b_{ext} (thick line) and model sulfate (dashed line). Model sulfate scale assumes 1100 m scale height. The dotted trace is proportional to the number of conservative quanta from a uniform emission grid remaining within the eastern United States, which defines a regional residence time for the airmass.

Global Pollution Modeling. (Patterson and Husar, 1981)

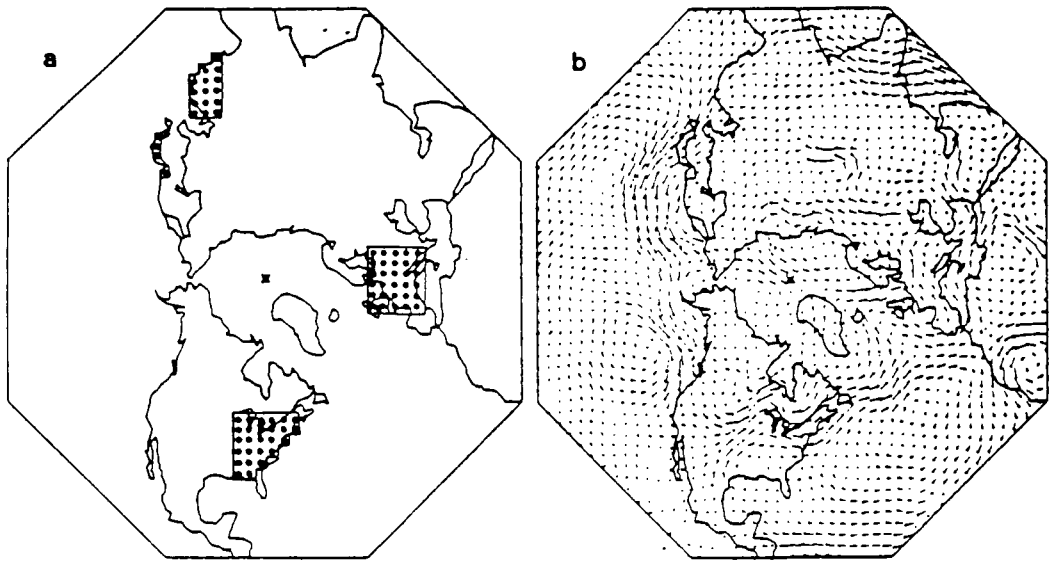


Fig. 5. (a) Emission field of trajectory origins. (b) Sample wind field grid.

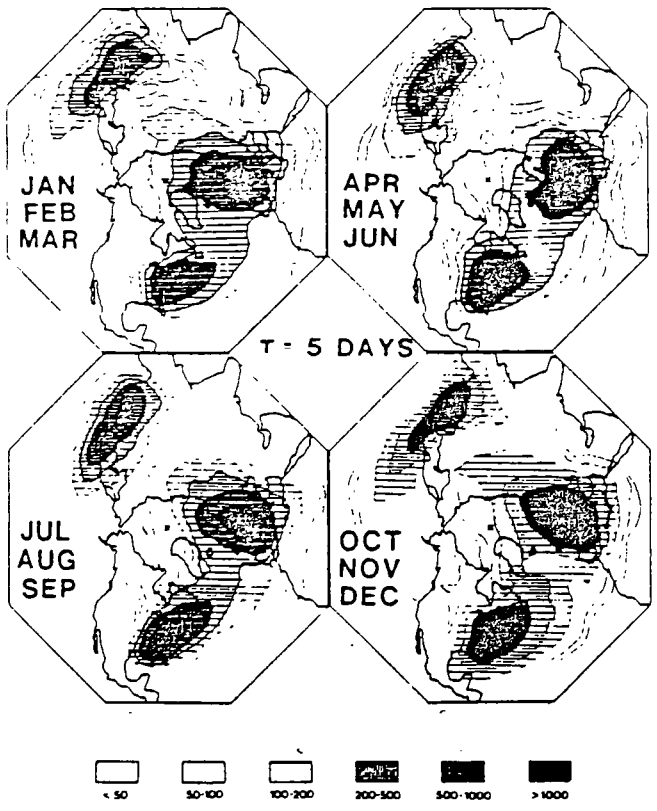


Fig. 5. Seasonal maps of vertical burden arising from 1974 850 mbar winds and 5-day residence time. Shadings represent sum of trajectory endpoints (puff arrivals per NMC grid square) weighted by decay for $\exp(-t/\tau)$.

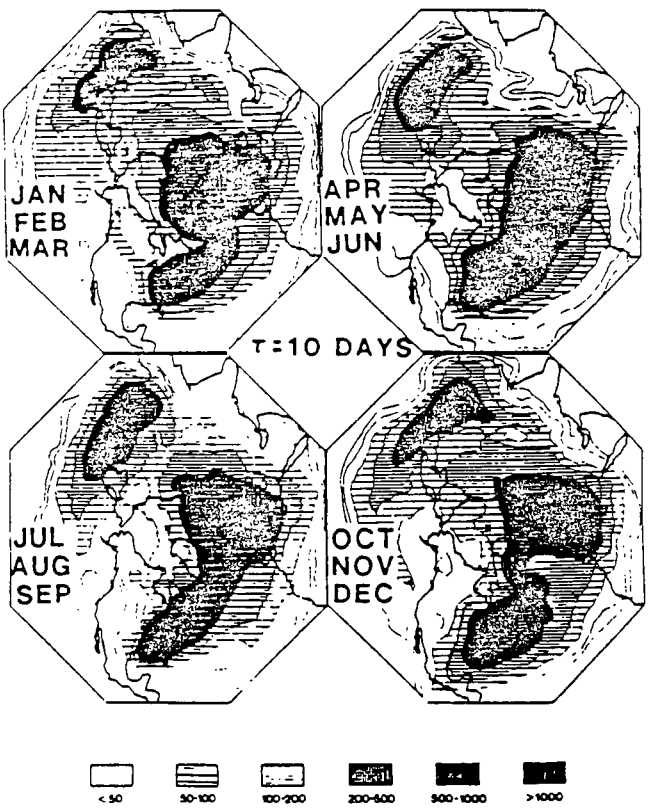
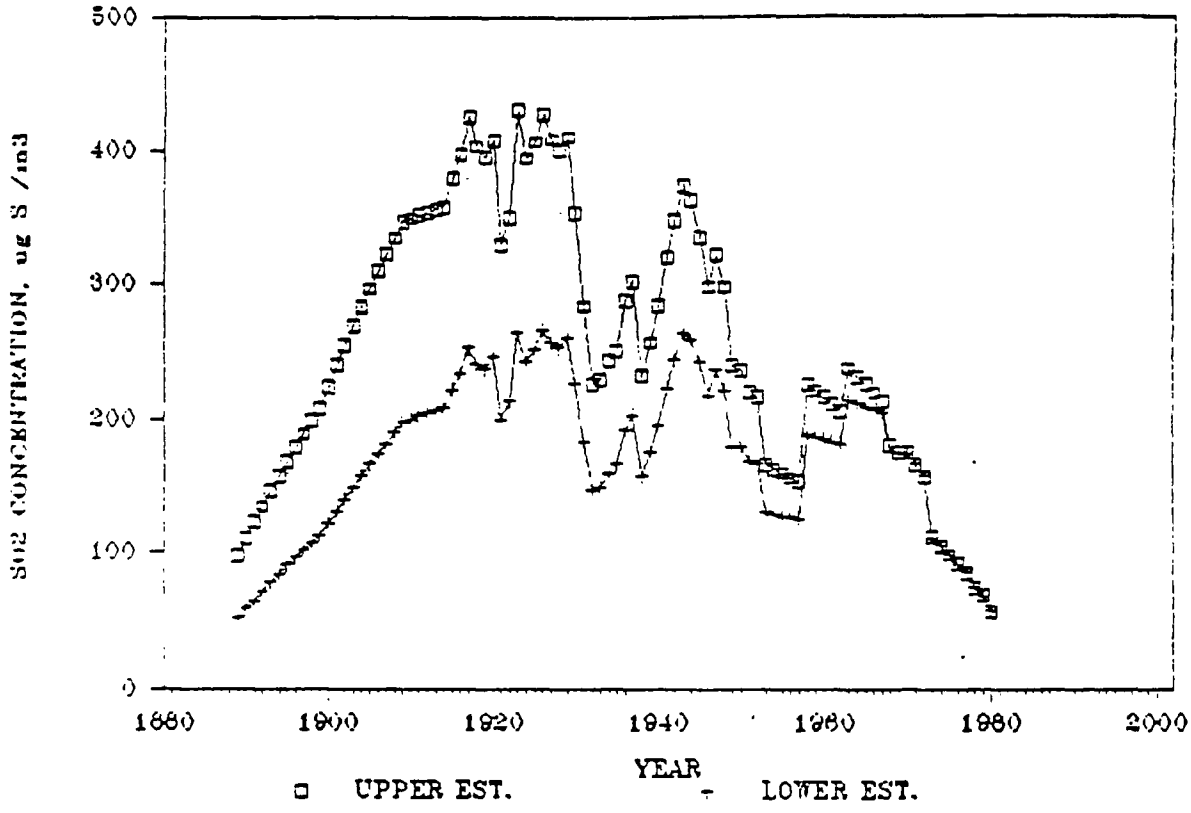


Fig. 3. Seasonal maps of vertical burden arising from 1974 850 mbar winds and 10-day residence time. Shadings represent sum of trajectory endpoints (puff arrivals per NMC grid square) weighted by decay factor $\exp(-t/\tau)$.

Retrospective Modeling. In this application, the model transfer matrices along with historical emission trends were used to reconstruct the SO_2 concentration trend in New York City Central Park for the period 1900-1980 (Husar et al., 1984) (Figure 6).



Estimated SO_2 concentration trend for New York city, Central Park.

1.7 APPROACH AND IMPLEMENTATION

This section states the approach and implementation of the proposed goals stated in section 1.3.

1.7.1 PC Based Version of the Monte Carlo Model (PCMC)

The new model implementation will operate on standard IBM compatible personal computers. While the model kernel will retain its features, the model will be completely wrapped into a graphic user interface. It will utilize the readily available Microsoft Windows graphic operating environment.

The model will be implemented using object oriented programming techniques using the C++ language.

1.7.2 Re-Calibration of the Model

Following the implementation, the PCMC model will be tested and its constants re-evaluated using more recent high quality aerosol databases. The candidate data sets include IMPROVE by the National Park Service; SCENES, a western U.S. research consortium, and the particle network of NESCAUM (Northeast States for Coordinated Air Use Management). These data sets will allow a more precise evaluation of the transformation and removal rate constants.

1.7.3 Development of Interfaces for Externally Generated Wind Fields

The previous model used NWS (National Weather Service) upper air and surface observations to derive the transport wind field. The gridded, x,y,z dependent wind vectors were generated by the Monte Carlo model itself.

In the PCMC model the above wind generation facilities will be preserved. In addition, "hooks" will be provided to allow the usage of externally generated model winds. Candidate wind grids include the NWS 100 km mesh predictive model that is available operationally. Another wind data source may be the MM4 Mesoscale Model by NCAR/Penn State.

The use of these external wind field will not eliminate the need to use the surface meteorological observations. Such input will provide estimates of solar radiation, precipitation events, relative humidity, and other variables required by the PCMC.

1.7.4 Present the PCMC Model Output Suitable for Policy Analysis

Unlike the first, research oriented version of the model, the PCMC will be oriented toward application in regulatory or other decision making. Hence, the output of the model will have to be tailored to answer questions relevant to the regulatory function of EPA or other agencies.

The first such regulatory activity under the 1990 Clean Air Act, is the formation of Transport Commissions. The charge of such commissions is to evaluate the regional (inter-state) aspects of air pollution. As its first application, The PCMC will provide the Transport Commissions with a tool to examine the regional air pollution transport from and to alternative source and receptor areas.

1.7.5 Interactive Graphic User Interface

The PCMC will be Windows-based program. It's operation is accomplished by menu selections and point and click queries. Programming knowledge will not be required.

An example application of the graphic user is outlined below for illustration purposes:

Suppose a member of the interstate Transport Commission wishes to evaluate the potential impacts of various sources on a given receptor region. The following user actions would be required.

- Select pollutant of concern. The emission field for that pollutant is automatically displayed on a map.
- Zoom in on the map and point to a specific location of interest. The program would automatically display a pie chart for the relative contribution various sources to that location.
- Point to specific sources on the map. This would be a query to retrieve all the characteristics of that source, including the emissions.
- From a menu ask for forward trajectories. The program would automatically draw the trajectories for the previously selected receptor location of a year, month, day or hour as specified be the user.
- Select "Show Monitoring Data" from a menu. The program would retrieve and display the available monitoring data for the previously specified location and pollutant.
- Select "Show Model Data" from a menu. The program would calculate instantly the model concentration pattern for the selected location and overlay it to the measured date. This would give the commission member a feel for the model performance.

The above illustration is but a small sample of the possible implementation for a user-friendly data interpretative model.

Since the data will be presented in physical units on maps and charts, and the user actions will be intuitive, the training and instruction time will be small compared to the use of the current modeling and data analysis software. Hypertext-based context-sensitive help will also be available to aid the user.

1.7.6 Use of Robust Software Building Blocks.

The PCMC will utilize modern, object oriented software building principles. It will be object oriented in principle as well as implementation. It will make use of "Software IC's" (integrated circuits) that are generic, robust, and suitable for integration into larger software applications.

These software building blocks will include Dynamic link Libraries (DLL's); Embedded Objects; Message based communication among objects; Software Construction sets, (such as ToolBook by Asymetrix Corporation. and Voyager data browser by Lantern Corporation.)

A key feature of object oriented approach is that most modules will be reusable. This will reduce the complexity, size and the maintenance cost of the software.

1.8 DELIVERABLES

The main deliverable of this project will be a PC based regional model based on the Monte Carlo principle. The model will be packaged as policy analysis tool, including tutorial, as well as on-line and hard copy documentation. The PCMC will be made available and distributable without royalty or other legal constraints.

Appendix 8

NAS review of Whitex - Limitations & Suggested Improvements

The Committee on Haze in National Parks and Wilderness Areas of the National Research Council, National Academy of Sciences prepared a report entitled "Haze in the Grand Canyon: An Evaluation of the Winter Haze Intensive Tracer Experiment" (WHITEX). The WHITEX experiment studied the effect of the Navajo Generating Station (NGS) upon visibility in Grand Canyon National Park (GCNP). The Environmental Protection Agency (EPA) is in the planning stage of a study (named Project MOHAVE) to determine the effects of the Mohave Generating Station on visibility in GCNP.

The NAS report on WHITEX noted a number of limitations in the study and some suggestions for how the study could have been improved. The purpose of this document is to identify how EPA intends to improve upon the limitations of WHITEX noted by the NAS and to incorporate the NAS suggestions into the Project MOHAVE study plan. NAS comments (paraphrased) on the WHITEX study that may be applicable to Project MOHAVE are listed below. The comments are followed by responses of how Project MOHAVE intends to consider these issues.

p.4 (Executive Summary) Committee identifies problems in the multiple linear regression analysis (DMB and TMBR):

1) Satisfactory tracers are not available for all major sources;

Response:

Project MOHAVE will attempt to identify tracers for all major sources, source types and source areas. For example, certain halocarbons may be used as tracers for the Los Angeles Basin. Also, sulfur to selenium ratios may be significantly different for different coal-fired powerplants. However, it is acknowledged that all major sources may not have satisfactory tracers

identified. This lack of complete source profiling often occurs in receptor modeling and does not necessarily preclude the use of receptor modeling to obtain quantitative results. However, uncertainties in source profiles needs to be incorporated into the error analysis.

2) Interpretation did not account for possible covariance between Navajo and other coal-fired powerplants in the area;

Response:

Trajectory analyses using the wind fields from the dynamic meteorological model will allow determination of times that the MPP plume and plumes from other sources are jointly present at receptor locations. This will facilitate consideration of covariance of impacts from MPP and other sources.

3) Both models treat sulfur conversion inadequately.

Response:

The exact methodologies of treating sulfur conversion in the receptor models has not yet been determined. Rather than scaling tracer by ambient surface relative humidity in TMBR, as in WHITEX, other methods will be considered. For example, data may be stratified into "wet" and "dry" conditions and the model run separately for each subset of data. Similarly for the DMB analysis, instead of assuming constant conversion rates for the entire data set, subsets of the data may be grouped, with constant rates over each group. It is acknowledged that some uncertainty in sulfur conversion is unavoidable; however, with the use of deterministic modeling, checked by tracer and sulfate data, along with receptor modeling, reasonable, quantitative estimates of sulfate contributions from each source may be obtained.

p.4 WHITEX did not quantitatively determine the fraction of SO_4 aerosol and resultant haze in GCNP attributable to NGS.

Response:

As discussed above and in response to other comments, with measurements, deterministic and receptor modeling and model reconciliation, quantitative apportionment of sulfate at GCNP can be done, within identified error bounds. After sulfate has been apportioned, statistical and first principle approaches can be used to attribute extinction.

p.4 WHITEX did not adequately quantify the sensitivity of the analysis to departure from model assumptions, nor did it establish an objective and quantitative rationale for selecting among various statistical models.
see response to 2nd comment on page 26.

p.4 The conceptual framework for DMB involved physically unrealistic simplifications for which the effect on quantitative assessments was not addressed.

Response:

As discussed elsewhere in the responses, more physically realistic assumptions will be made wherever possible. However some simplifications will remain, as in all modeling studies. The effect of variations in assumptions can be studied to some extent with sensitivity analysis. Also, comparison to deterministic models (which also contain simplifications) may help determine the effect of simplifications upon quantitative assessments.

p.4 The data base contained weaknesses; especially important was the lack of measurements below the rim and the paucity of background measurements (particularly SO_4).

Response:

The conceptual plan for Project MOHAVE calls for monitoring below the rim of the Grand Canyon and increased background monitoring compared to WHITEX, including SO_4 and tracer. It should be recognized that the number of feasible monitoring sites is limited due to power requirements and

the inaccessibility of some areas.

p.4 The background measurements were inadequately incorporated into the data analyses (in particular, SO_4).

Response:

The NAS comments emphasize that not enough sampling sites were located in the vicinity of GCNP (p. 25). In addition, tracer was not measured at many locations and only a small subset of tracer data were analyzed. Project MOHAVE will have more sampling sites in the vicinity of GCNP and operation over a 12-15 month period, compared to 6 weeks for WHITEX. This includes more sulfate sample analysis and far more sample analysis of tracer. However, due to accessibility problems and power requirements, the number of feasible sampling sites in the location of GCNP is limited. Thus, the actual number and location of sites may be less than ideal.

p.20 Literature does not demonstrate that MLR can successfully apportion secondary species among several source types; therefore is not advisable to rely solely on such models for the success of a major field experiment.

Response:

Project MOHAVE is emphasizing the use of deterministic models rather than MLR for apportionment of secondary species. The analysis will also use receptor models and eigenvector analysis as a check of the deterministic models.

p.21 Deterministic met. modeling did not reproduce the diurnal fluctuation in wind flow observed at Page.

see response to next comment

p.21 The met. data and deterministic meteorological modeling do not allow quantification of the contribution that NGS might have made to haze at GCNP. The deterministic modeling cannot pinpoint the location of the NGS

plume nor its entrainment into the canyon. The model uses a grid size of 5 km; hence it cannot reproduce the complex topography of GCNP nor the associated small scale meteorological effects, such as gravity flows. Thus the meteorological studies provide only qualitative evidence of transport.

Response:

Project MOHAVE will more thoroughly model MPP using increased meteorological data and greater resolution of topography. Modeling will be done for the entire 12-15 month study period. Wind profilers will provide a much increased meteorological data base compared to the WHITEX study. Model grid size will be 500 m at key areas, allowing greater topographic resolution and improved representation of small scale flows. It should be understood that it is impossible to exactly model wind fields; of particular difficulty is flow in highly complex terrain such as the study area. Monitoring and modeling of moisture and chemical transformations will allow for a reasonable quantification of MPP impacts to haze at GCNP.

p. 24 No tracer was used in WHITEX to evaluate urban emissions; therefore the fraction of haze attributable to these sources is impossible to calculate.

Response:

Tracers for urban areas will be investigated. For example, certain halocarbons have been identified as tracers for the Los Angeles Basin. Other urban areas, particularly Las Vegas will also be investigated for endemic tracers. In addition, the deterministic modeling will identify the time periods when emissions from urban areas are in the Grand Canyon area.

p. 24 The source profile for powerplants was based on limited aircraft measurements of NGS emissions downwind from the stacks. The copper smelter profile was based on old and uncertain data from the literature.

Response:

The planners of Project MOHAVE are aware of the critical nature of accurate source profiles for use in receptor models. All available data for

powerplant emissions in the region will be used to generate powerplant source profiles. The most recent data for smelter emissions will be used. Resource limitations preclude significant field efforts to document source profiles of all important sulfur sources with the potential to impact the GCNP area.

p. 24 Variabilities and uncertainties in NGS CD4 emission rates led to substantial uncertainties in the day to day relationship between CD_4 and NGS sulfur emissions.

Response:

Unlike the WHITEX study, which used tracer data mainly for receptor modeling, Project MOHAVE will also use tracer for estimating plume dilution factors. For this purpose, a constant tracer release rate is desirable. With variations in MPP load, this will result in variations in tracer to sulfur ratios. For use in receptor modeling, as in WHITEX tracer concentrations need to be scaled to the sulfur emissions, which requires plume age. The more sophisticated meteorological modeling to be done for Project MOHAVE will give a better calculation of plume age than the simple trajectory models used in WHITEX.

p. 24 At Hopi Point, CD_4 concentrations were determined for only 36 samples, an undesirably small data set for the types and large numbers of statistical analyses performed on the data.

Response:

The WHITEX study analyzed a small number of samples of CD_4 because of the very high analysis costs. It is expected that perfluorocarbons will be used for Project MOHAVE, for which the analysis costs are not prohibitive. The tracer sample size will be many times the size for WHITEX, allowing for a sufficiently large data set for use in statistical analyses.

p. 24 The ratio of SO_2 to CD_4 in the stack was not analyzed.

Response:

Project MOHAVE intends to analyze some stack samples for SO_2 to tracer ratio.

p. 24 The report provides little documentation of procedures and quality assurance for the sampling and analysis of ambient CD_4.

Response:

The participants in Project MOHAVE are acutely aware of quality assurance problems with some past tracer experiments. The skepticism regarding the quantitative use of tracers requires not only careful quality assurance, but also detailed documentation of the procedures and quality assurance performed. Project MOHAVE reports will provide detailed documentation of quality assurance for tracer and other data collection.

p. 25 Without data from more stations, the effect of NGS emissions is difficult to differentiate from other sources in the region.

Response:

Project MOHAVE expects to have data from additional stations in the area of GCNP compared to WHITEX. Perhaps more significantly, the deterministic modeling will help differentiate impacts from MPP and other sources.

p. 26 WHITEX design did not provide the data necessary to quantify the effects of departures from the statistical assumptions made.

Response:

see response to comment #6, page 4.

p. 26 SO_4 contribution attributed to NGS depends strongly on the model chosen, the tracers included in the model, and the criteria by which the model is fit to the data. To establish a more rational basis for quantitative attribution, more attention must be given to alternative formulations of TMBR and DMB and the criteria for selecting among them. However, even if these criteria were adequately considered, the statistical results would most likely remain non-robust in the sense that source attributions generated by the various statistical models would probably still differ substantially from one another. One difficulty is that the number of plausible alternative models is substantial relative to the number of samples for which CD_4 data are available. As the number of models increase, so does the likelihood that one

of them will test significant merely by chance.

Response:

Model formulations will be done based on theoretical considerations. Sensitivity analysis of varying model assumptions within reasonable ranges will be done to determine the bounds of possible results. It is possible that different receptor (and deterministic) models will yield significantly different results. Reconciliation of model results will be done at this point. Many more samples of tracer will be available compared to the WHITEX study, thus decreasing the likelihood that a model will test significant by chance.

p. 26 WHITEX assumed SO_4 yields from NGS and smelter emissions were proportional to ambient relative humidity. This is a simple and indirect assumption, which scales intermittent processes along the entire trajectory at cloud level directly to a continuous variable measured at ground level.

Response:

In Project MOHAVE, the effect of moisture upon sulfate formation will be treated more rigorously than done in the WHITEX report. In addition to surface humidity measurements, the deterministic meteorological model will give estimates of humidity at many vertical levels. This information will include prediction of clouds, which can be compared to satellite observations. Rather than scaling linearly with relative humidity, a determination will be made whether or not the plume is in contact with clouds. Rates of sulfate formation are thought to occur rapidly in clouds, and quite slowly without, particularly in winter. Thus, stratification of data into "wet" and "dry" categories seems appropriate.

p. 31 Given the overriding importance of the RH scaling factor, the committee believes that the sensitivity of results to alternative assumptions should have been explored in formulating the models used for the TMBR and DMB analyses. The NPS WHITEX report assumes that the contribution of background sources, such as other power plants and urban areas, were unaffected by RH. The committee believes the report should have considered the possibility that yields from other sources were also affected by RH.

Response:

It is likely that contributions from other sources are affected by relative humidity. This will be considered in the analysis.

p. 31 The DMB analyses are dependent on unique "plume ages", the validity of which is questionable. Plume ages were estimated only for NGS and not for other sources.

Response:

Plume ages will be estimated for MPP and a variety of other sources using wind fields generated by the dynamic meteorological model; this should provide reasonable estimates of plume ages.

p. 31 DMB is based upon linear models for the oxidation of SO_2 to SO_4 and for the deposition of SO_2 and SO_4. In reality, both processes are likely to occur at rates that can vary greatly in space and time.

Response:

see response to comment 3, page 4.

p. 32 Nonuniformities in conversion and deposition rates lead to variabilities in the relationship between SO_4 concentrations measured at the receptor sites and tracer concentrations used in the regression analyses. Because these nonuniformities were not taken into account in the DMB formulation, the DMB results are of questionable applicability.

Response:

see response to comment 3, page 4.

p. 32 Possible covariance of impacts from NGS and other coal-fired power plants makes it difficult to statistically distinguish the relative effects of NGS and other plants.

Response:

Trajectory analyses using the wind fields from the dynamic meteorological model will allow determination of times that the MPP plume and plumes from other sources are jointly present at receptor locations. This will facilitate consideration of covariance of impacts from MPP and other sources.

p. 35 No H_2O_2 measurements were made at or near GCNP during WHITEX.

Response:

Measurements of H_2O_2 will be made in the study area, under varying conditions.

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Appendix 9

SURVEY OF A VARIETY OF RECEPTOR MODELING TECHNIQUES

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Abstract

The chemical mass balance (CMB) formalism has been used on a semi-routine basis to apportion emissions used to mass concentrations at specific receptor sites. Recently, two other techniques, differential mass balance (DMB) and tracer mass balance regression (TMBR) have been used to apportion secondary aerosols to sources and source types of a variety of receptor areas. CMB uses known source and receptor measured tracer profiles (gradients in tracer concentration at one point in time) to apportion sources at one point in time. DMB uses gradients in trace elements across space, while TMBR uses changes in tracers across time to achieve apportionment of primary as well as secondary aerosol species. Assumptions and limitations of each approach will be addressed and a unified formalism building on strengths of all three approaches will be presented.

SURVEY OF A VARIETY OF RECEPTOR MODELING TECHNIQUES

Introduction

Receptor modeling approaches rely on known physical and chemical characteristics of gases and particles at receptors and sources to attribute aerosols to a source or source type. Historically, the CMB formalism has been used to apportion primary particles. This formalism uses known relationships between emitted tracers and an assumption that various tracer profiles stay constant as material is transported from source to receptor. These tracer profiles are then used to apportion primary species for each time period that a measurement is made at a receptor site. Other common types of models include principal component analysis (PCA) and multiple linear regression (MLR). Explanations of these models are given by Watson[1,2],[3] Chow,[4] and Hopke.[5] All these models are special cases of a General Mass Balance (GMB) model which is deterministic in nature. A regressional model similar to MLR is derivable from the GMB equations and will be referred to as the TMBR model. The TMBR model incorporates changes in tracer material over time to apportion both primary and secondary aerosols. Finally, the DMB model, a special case of GMB and referred to here as a receptor oriented model, is really a hybrid model in that it relies on tracer material to establish atmospheric dispersion characteristics but deterministically accounts for deposition and oxidation. Stevens and Lewis,[6] Lewis and Stevens,[7] and Dzubay et al.,[8] have used models similar to the TMBR and GMB to create a hybrid model which they have used for source apportionment.

General Mass Balance Equations

Each special case of GMB has its own set of limiting assumptions and special requirements for solution. The assumptions that need to be satisfied for the mathematical model to be valid will be apparent during the process of derivation of the model equations. Nevertheless, the assumptions will be explicitly stated after the derivations of the model equations have been explained. The statistical aspects of the estimation of the fractional contribution by a given source and the calculation of the associated uncertainties will also be presented.

Notational Conventions

The following notation will be used throughout.

Total number of species under consideration = m.
Total number of sources under consideration = n.
Total number of sampling periods = s.

The subscript i will be used for indexing the species, j for sources and k for sampling periods.

c_{ijk} = concentration of aerosol species i at source j corresponding to sampling period k.

C_{ijk} = concentration of aerosol species i at the receptor, attributable to source j corresponding to sampling period k.

t_{jk} = travel time for the air mass from source j to the receptor, corresponding to sampling period k.

r_{ijk} = a factor that accounts for deposition of aerosol species i from source j, for sampling period k.

r^*_{ijk} = a factor that accounts for the formation of aerosol species i from a parent species i^* emitted by source j as well as its deposition during transport for sampling period k.

d_{jk} = a factor that accounts for dispersion of aerosol mixture from source j during sampling period k, as the mixture travels from the source to the receptor.

Whenever a subscript i denotes a secondary aerosol species, then the subscript i^* will denote the corresponding parent aerosol species. For instance, if i denotes SO_4 then i^* will stand for SO_2.

Model Equations

It follows from the definitions that for primary aerosol species

$$C_{ijk} = c_{ijk} r_{ijk} d_{jk} \tag{1}$$

and for secondary aerosol components we have

$$C_{ijk} = c_{ijk} r_{ijk} d_{jk} + c_{i^*jk} r^*_{ijk} d_{jk}. \tag{2}$$

The quantities r_{ijk} are a function of deposition rates and transport time while r^*_{ijk} are functions of deposition and transport times as well as conversion rates.

Simple functional forms for r_{ijk} and r^{*}_{ijk} can be derived if it is assumed that chemical conversion and deposition are governed by first order mechanisms and conversion and deposition rates are constant in space over some finite increment in time.

Let $X(t)$ denote the mass, at time t after emission, of a species i in a unit volume of aerosol mixture. Assume, ignoring dispersion temporarily,

$$\frac{dX(t)}{dt} = -(K_c + K_1)X(t) \tag{3}$$

which, when solved, yields

$$X(t) = X(0)exp(-(K_c + K_1)t) \tag{4}$$

where $X(0)$ is the mass at time 0 in unit volume of aerosol mixture, i.e. the concentration of the species at the source. The quantities K_c and K_1 are the conversion and deposition rates, respectively, for the species under consideration. The conversion and deposition rates have been assumed to remain constant throughout the transport path in space and time. If $d(t)$ denotes the dispersion factor corresponding to t time units after emission of the aerosol mixture, then

$$X(t)x = X(0)exp(-(K_c + K_1)t)d(t). \tag{5}$$

The factor accounting for conversion and deposition thus has the form $exp(-(K_c + K_1)t)$ in this case.

Suppose $Y(t)$ is the concentration of a secondary aerosol species at time t after the parent species i is emitted by the source. Let $X(t)$ be the concentration of the parent species at time t after emission. If the dispersion factor is temporarily ignored the following pair of differential equations hold:

$$\frac{dY(t)}{dt} = K_c X(t) - K_2 Y(t) \tag{6}$$

and

$$\frac{dX(t)}{dt} = -(K_c + K_1)X(t) \tag{7}$$

where K_2 refers to the deposition rate of the secondary aerosol which is assumed to be nonconverting. Once again, it has been assumed that the deposition and conversion parameters are constant throughout the transport path in space and time. Solution of the pair of differential equations yields the relation

$$Y(t) = \frac{K_c}{K_1 + K_c - K_2}\{exp(-K_2 t) - exp(-(K_1 + K_c)t)\}. \tag{8}$$

If now the dispersion factor $d(t)$ is taken into account

$$Y(t) = \frac{K_c}{K_1 + K_c - K_2}\{exp(-K_2 t) - exp(-(K_1 + K_c)t)\}d(t). \qquad (9)$$

From this relation it becomes evident that the factor accounting for the formation of the secondary aerosol species from its parent species as well as its deposition during transport is of the form

$$\frac{K_c}{K_1 + K_c - K_2}\{exp(-K_2 t) - exp(-(K_1 + K_c)t)\}. \qquad (10)$$

Based on the above arguments, when the conversion and deposition rates of the various species remain constant throughout the duration of transport from the source to the receptor

$$r_{ijk} = exp(-(K_c(i,j,k) + K_d(i,j,k))t_{jk}) \qquad (11)$$

and

$$r^*_{ijk} = \frac{K_c(i^*,j,k)}{K_c(i^*,j,k) + K_d(i^*,j,k) - K_d(i,j,k)} \times$$
$$\{exp(-K_d(i,j,k)t_{jk}) - exp(-[K_c(i^*,j,k) + K_d(i^*,j,k)]t_{jk})\} \qquad (12)$$

where

$K_c(i,j,k)$ = conversion rate of species i from source j to its secondary form, during sampling period k.

$K_d(i,j,k)$ = deposition rate of species i from source j during sampling period k.

Let C_{ik} = concentration of aerosol component i at the receptor during sampling period k. Since the concentration of aerosol component i at the receptor is the sum of the concentrations attributable to various sources, the mass balance equation becomes

$$C_{ik} = \sum_{j=1}^{n} C_{ijk} \qquad (13)$$

for each sampling period $k = 1, 2, \ldots, s$. From this basic equation various special cases can be derived.

CMB Model

The first special case of the GMB equations to be examined is the Chemical Mass Balance formalism.

Model Equations

Suppose our list of aerosol components includes only material that is nonreactive and maintains relative ratios between various species as material is transported from source to receptor. In this case $K_c(i,j,k)$ are all zero and $K_d(i,j,k)$ are the same for all elements i. Their common value is denoted by $K_d(j,k)$ indicating the nondependence on i. This implies that the quantities r_{ijk} do not depend on i. Then,

$$\frac{C_{ijk}}{C_{i'jk}} = \frac{c_{ijk}r_{ijk}d_{jk}}{c_{i'jk}r_{i'jk}d_{jk}} = \frac{c_{ijk}}{c_{i'jk}} \tag{14}$$

which implies that the signature for source j at the source equals the signature for source j as perceived at the receptor.

Let $S_{jk} = \sum_{i=1}^{m} C_{ijk}$. The quantity S_{jk} is the concentration of the aerosol mixture at the receptor during sampling period k that is attributable to source j. The fraction a_{ijk} defined by

$$a_{ijk} = \frac{C_{ijk}}{S_{jk}} \tag{15}$$

is then the fraction of species i in the aerosol mixture at the receptor attributable to source j during sampling period k. Assuming Equation (14) is valid, the numbers a_{ijk} for $i = 1, 2, \ldots, m$ represent the source signature for source j for sampling period k. From Equations (13) and (15) it follows that the set of Equations in (16) below also holds.

$$C_{ik} = \sum_{i=1}^{n} a_{ijk}S_{jk} \tag{16}$$

If the a_{ijk} for all the sources affecting the receptor sites are known, then 16 is a system of linear simultaneous equations in n unknowns $S_{1k}, S_{2k}, \ldots, S_{nk}$, for each of the sampling periods $k = 1, 2, \ldots, s$. These are in fact the chemical mass balance equations. The rank of the system of equations for each k must be equal to n in order to uniquely solve these equations. In particular, the numbers of equations must be greater than or equal to the number of chemical species (i).

Solutions to the CMB equations that have been used are: 1) a tracer solution; 2) a linear programming solution; 3) an ordinary weighted least squares solution with or without an intercept; 4) a ridge regression weighted least squares solution with or without an intercept; and 5) an effective variance least squares solution with or without an intercept. An estimate of the uncertainty associated with the source contributions is an integral part of several of these solution methods.

Weighted linear least squares solutions are preferable to the tracer and linear programming solutions because: 1) theoretically they yield the most likely solution to the CMB equations providing model assumptions are met; 2) they can make use of all available chemical measurements, not just the so- called tracer species; 3) they are capable of analytically estimating the uncertainty of the source contributions;

CMB software in current use[9] applies the effective variance solution developed and tested by Watson[11] because this solution: 1) provides realistic estimates of the uncertainties of the source contributions (owing to its incorporation of both source profile and receptor data uncertainties); and 2) chemical species with higher precisions in both the source and receptor measurements are given greater influence than are species with lower precisions. The effective variance solution is derived[10] by minimizing the weighted sums of the squares of the differences between the measured and calculated values of C_{ik} and a_{ij}. The solution algorithm is an iterative procedure which calculates a new set of S_{jk} based on the S_{jk} estimated from the previous iteration.

Watson[12] found that individual sources with similar source profiles would yield unreliable values if included in the same chemical mass balance. Henry[13] proposed a quantitative method of identifying this interference between this similar source compositions, which is known as "collinearity." He uses the "singular value decomposition" define an "estimable space into which resolvable sources should lie." The sources which do not fall into this estimable space are collinear, or too similar to be resolved from the sources which do lie within the estimable space.

Williamson and Dubose[14] claimed that the ridge regression reduces colinearities. Henry[13] tested the ridge regression solution with respect to the separation of urban and continental dust and found that the bias resulted in physically unrealistic negative values for several of the a_{ij}. The ridge regression solution has not been used in the CMB since these tests were published.

CMB Model Assumptions

The CMB model assumptions are:

- Compositions of source emissions are constant over the period of ambient and source sampling.
- Chemical species do not react with each other, i.e., they add linearly.

- All sources with a potential for significantly contributing to the receptor have been identified and have had their emissions characterized.
- The sources' compositions are linearly independent of each other.
- The number of sources or source categories is less than or equal to the number of chemical species.
- Measurement uncertainties are random, uncorrelated, and normally distributed.

Effects of Deviations from CMB Model Assumptions

Assumptions 1 through 6 for the CMB model are fairly restrictive and will never be totally complied within actual practice. Fortunately, the CMB model can tolerate reasonable deviations from these assumptions, though these deviations increase the stated uncertainties of the source contribution estimates.

The CMB model has been subjected to a number of tests to determine its abilities to tolerate deviations from model assumptions.[3, 12, 13, 15, 16, 17, 18, 19, 20, 21, 22] These studies all point to the same basic conclusions regarding deviations from the above-stated assumptions.

With regard to Assumption 1, source compositions, as seen at the receptor, are known to vary substantially among sources, and even within a single source over an extended period of time. These variations are both systematic and random and are caused by three phenomena: 1) transformation and deposition between the emission point and the receptor; 2) differences in fuel type and operating processes between similar sources or the same source in time; and 3) uncertainties or differences between the source profile measurement methods. Evaluation studies have generally compared CMB results from several tests using randomly perturbed input data and from substitutions of different source profiles for the same source type. The general conclusions drawn from these tests are:

- The error in the estimated source contributions due to biases in all of the elements of a source profile is in direct proportion to the magnitude of the biases.
- For random errors, the magnitude of the source contribution errors decreases as the number of components increases.

The most recent and systematic tests are those of Javitz[22] which apply to a simple four-source urban airshed and a complex ten-source urban airshed. These tests, with 17 commonly measured chemical species, showed that primary mobile, geological, coal-fired power plant, and vegetative burning source types can be apportioned with uncertainties of approximately 30% when coefficients of variation in the source profiles are as high as 50%. This performance was demonstrated even without the presence of unique "tracer" species such as selenium for coal-fired power plants or soluble potassium for vegetative burning. In a complex urban airshed, which added residual oil combustion, marine aerosol, steel production, lead smelting, municipal incineration, and a continental background aerosol, it was found that the geological, coal-fired power plant, and background source profiles were collinear with the measured species. At coefficients of variation in the source profiles as low as 25%, average absolute errors were on the order of 60%, 50%, and 130% for the geological, coal-burning, and background sources, respectively. All other sources were apportioned with average absolute errors of approximately 30% even when coefficients of variation in the source profiles reached 50%. Once again, these tests were performed with commonly measured chemical species, and results would improve with a greater number of species which are specifically emitted by the different source types.

With regard to the nonlinear summation of species, Assumption 2, no studies have been performed to evaluate deviations from this assumption. While these deviations are generally assumed to be small, conversion of gases to particles and reactions between particles are not inherently linear processes. This assumption is especially applicable to the end products of photochemical reactions and their apportionment to the sources of the precursors. Further model evaluation is necessary to determine the tolerance of the CMB model to deviations from this assumption. The current practice is to apportion the primary material which has not changed between source and receptor. The remaining quantities of reactive species such as ammonium, nitrate, sulfate, and elemental carbon are then apportioned to chemical compounds rather than directly to sources. While this approach is not as satisfying as a direct apportionment, it at least separates primary from secondary emitters and the types of compounds apportioned give some insight into the chemical pathways which formed them. As chemical reaction mechanisms and rates, deposition velocities, atmospheric equilibrium, and methods to estimate transport and aging time become better developed, it may be possible to produce "fractionated" source profiles which will allow this direct attribution or reactive species to sources. Such apportionment will require measurements of gaseous as well as particulate species at receptor sites.

A major challenge to the application of the CMB is the identification of the primary contributing sources for inclusion in the model, Assumption 3. Watson[12]

systematically increased the number of sources contributing to his simulated data from four to eight contributors while solving the CMB equations assuming only four sources. He also included more sources in the least squares solutions than those which were actually contributors, with the following results:

- Underestimating the number of sources had little effect on the calculated source contributions if the prominent species contributed by the missing sources were excluded from the solution.
- When the number of sources was underestimated, and when prominent species of the omitted sources were included in the calculation of source contributions, the contributions of sources with properties in common with the omitted sources were overestimated.
- When source types actually present were excluded from the solution, ratios of calculated to measured concentrations were often outside of the 0.5 to 2.0 range, and the sum of the source contributions was much less than the total measured mass. The low calculated/measured ratios indicated which source compositions should be included.
- When the number of sources was overestimated, the sources not actually present yielded contributions less than their standard errors if their source profiles were significantly distinct from those of other sources. The over-specification of sources decreased the standard errors of the source contribution estimates.

Recent research suggests that Assumption 3 should be restated to specify that source contributions above detection limits should be included in the CMB. At this time, however, it is not yet possible to determine the "detection limit" of a source contribution at a receptor since this is a complicated and unknown function of the other source contributions, the source composition uncertainties and the uncertainties of the receptor measurements. Additional model testing is needed to define this "detection limit."

The linear independence of source compositions required by Assumption 4 has become a subject of considerable interest since the publication of Henry's[13] singular value decomposition (SVD) analysis. As previously noted, this analysis provides quantitative measures of collinearity and the sensitivity of CMB results to specific receptor concentrations. These measures can be calculated analytically in each application. Henry[13] also proposed an optimal linear combination of source contributions that have been determined to be collinear.

Other "regression diagnostics" have been summarized by Belsley[23] and have been applied to the CMB by DeCesar.[19, 20] Kim and Henry[24] show that most of these diagnostics are useless because they are based on the assumption of zero uncertainty in the source profiles. They demonstrate, through the examination of randomly perturbed model input data, that the values for these diagnostics vary substantially with typical random changes in the source profiles.

Tests performed on simulated data with obviously collinear source compositions typically result in positive and negative values for the collinear source types as well as large standard errors on the collinear source contribution estimates. Unless the source compositions are nearly identical, the sum of these large positive and negative values very closely approximates the sum of the true contributions.

With most commonly measured species (e.g., ions, elements and carbon) and source types (e.g., motor vehicle, geological, residual oil, sea salt, steel production, wood burning and various industrial processes), from five to seven sources are linearly independent of each other in most cases.[22]

Gordon[15] found instabilities in the ordinary weighted least square solutions to the CMB equations when species presumed to be "unique" to a certain source type were removed from the solution. Using simulated data with known perturbations ranging from 0 to 20 percent, Watson[12] found: "In the presence of likely uncertainties, sources such as urban dust and continental background dust cannot be adequately resolved by least squares fitting, even though their compositions are not identical. Several nearly unique ratios must exist for good separation."

With regard to Assumption 5, the true number of individual sources contributing to receptor concentrations is generally much larger than the number of species that can be measured. It is therefore necessary to group sources into source types of similar compositions so that this assumption is met. For the most commonly measured species, meeting Assumption 4 practically defines these groupings.

With respect to Assumption 6 (the randomness, normality, and the uncorrelated nature of measurement uncertainties), there are no results available from verification or evaluation studies. Every least squares solution to the CMB equations requires this assumption, as demonstrated by the derivation of Watson.[11] In reality, very little is known about the distribution of errors for the source compositions and the ambient concentrations. If anything, the distribution probably follows a log-normal rather than a normal distribution. Ambient concentrations can never be negative, and a normal distribution allows for a substantial proportion of negative values, while a log-normal distribution allows no negative values. For small errors (e.g., less than 20%), the actual distribution may not be important, but for large errors, it probably is important. A symmetric distribution becomes

less probable as the coefficient of variation of the measurement increases. This is one of the most important assumptions of the solution method that requires testing.

Model Input and Output Data

The chemical mass balance modeling procedure requires: 1) identification of the contributing sources types; 2) selection of chemical species to be included; 3) estimation of the fraction of each of the chemical species which is contained in each source, i.e., the source compositions); 4) estimation of the uncertainty in both ambient concentrations and source compositions; and 5) solution of the chemical mass balance equations, and 6) validation and reconciliation. Each of these steps requires different types of data.

Emissions inventories are examined to determine the types of sources which are most likely to influence a receptor. Principal components analysis applied to a time series of chemical measurements is also a useful method of determining the number and types of sources. After these sources have been identified, profiles acquired from similar sources[25] (identify most of the available source profiles) are examined to select the chemical species to be measured. Watson[12] demonstrates that the more species measured, the better the precision of the CMB apportionment.

The ambient concentrations of these species, C_i, and their fractional amount in each source-type emission, F_{ij}, are the measured quantities which serve as CMB model input data. These values require uncertainty estimates, σ_{Ci} and σ_{Fij}, which are also input data. Input data uncertainties are used both to weight the importance of input data values in the solution and to calculate the uncertainties of the source contributions. The output consists of: 1) the source contribution estimates (S_j) of each source type; 2) the standard errors of these source contribution estimates. 3) the amount contributed by each source type to each chemical species.

TMBR Model

The TMBR model is a multiple regression based model which may be used to apportion an aerosol species of interest measured at a receptor site to the various contributing sources. The actual regression analysis may be performed using the method of ordinary least squares. However, since the independent variables in this model are ambient concentrations of various aerosol components which are measured with error, the method of Orthogonal Distance Regression (ODR) is ex-

pected to give better estimates of the source contributions. A detailed theoretical discussion of the method of ODR may be found in the book by Fuller (1987).[26]

Model Equations

In this section it is shown that, under appropriate assumptions, the general mass balance model can be reduced to a simpler linear model. Let aerosol component $i = 1$ be a secondary aerosol with $i^* = 2$ denoting the corresponding parent species. It is of interest to determine the fractional contribution to the ambient concentrations of this secondary aerosol component by a distinguished source which will be denoted by the subscript $j = 1$. We will also assume that aerosol species i_1 is a tracer for this distinguished source. Let sources $j = 2$ thru $j = n_2$ have an associated tracer species i_2, sources $j = n_2 + 1$ thru $j = n_3$ have an associated tracer species i_3 etc., and sources $j = n_{h-1} + 1$ thru $j = n_h$ have an associated tracer i_h. Sources $j = n_h + 1$ thru $j = n$ may be unknown sources or may be known sources with tracers that are not measured at the receptor. For the sake of uniformity of notation we let $n_1 = 1$. Thus the n sources have been partitioned into $h+1$ groups, each of the first h groups of sources being associated with a unique tracer species or with a fraction of some reference species that has been calculated using CMB or some other appropriate model.

In general for $1 \leq u \leq h$ and $n_{u-1} + 1 \leq j \leq n_u$ we have

$$C_{1jk} = r^*_{1jk} d_{jk} c_{2jk} = \frac{r^*_{1jk}}{r_{i_u jk}} \frac{c_{2jk}}{c_{i_u jk}} C_{i_u jk} = \beta_{i_u jk} C_{i_u jk} \tag{17}$$

Therefore,

$$\sum_{j=n_{u-1}+1}^{n_u} C_{1jk} = \sum_{j=n_{u-1}+1}^{n_u} \beta_{i_u jk} C_{i_u jk} = \beta_{i_u k} C_{i_u k} \tag{18}$$

where $\beta_{i_u k}$ is defined as

$$\beta_{i_u k} = \frac{\sum_{j=n_{u-1}+1}^{n_u} \beta_{i_u jk} C_{i_u jk}}{C_{i_u k}} \tag{19}$$

For $n_h + 1 \leq j \leq n$ let

$$\beta_{0k} = \sum_{j=n_h+1}^{n} C_{1jk}. \tag{20}$$

The general mass balance equation then reduces to the equation

$$C_{1k} = \beta_{0k} + \sum_{u=1}^{h} \beta_{i_u k} C_{i_u k} \tag{21}$$

for each sampling period $k = 1, 2, \ldots, s$.

If the quantities $\beta_{i_u k}$ are all independent of k for each u, $\beta_{i_u k} = \beta_{i_u}$, and the above set of equations reduce to

$$C_{1k} = \beta_0 + \sum_{u=1}^{h} \beta_{i_u} C_{i_u k} \tag{22}$$

The quantities $C_{i_u k}$ are ambient concentrations of the tracer species $i_1, i_2, \ldots, i_h$ and are assumed known. The quantities C_{1k} are the ambient concentrations of the aerosol species being apportioned and are also assumed known. We thus have a set of s linear equations in $h+1$ unknowns $\beta_0, \beta_{i_1}, \beta_{i_2}, \ldots, \beta_{i_h}$. If the system of equations has rank $h+1$, then these unknown beta coefficients may be obtained by solving the above system of linear equations. The apportionment of the species of interest to the various groups of sources is then carried out by calculating the individual terms of the equations above.

In certain instances it is known that the beta coefficients will differ significantly from one time period to another. In such cases it may be possible to determine, based on physical and chemical reasons, a function of the field measurements, the sampling period and the source, which we denote by ϕ_{jk}, such that it is more reasonable to assume the quantities $\beta_{i_u k}/\phi_{jk}$ are constant for all sampling periods rather than the quantities $\beta_{i_u k}$. In such cases we define $\gamma_{i_u} = \beta_{i_u k}/\phi_{jk}$. For uniformity of notation we define γ_0 to be equal to β_0. This results in the system of linear equations

$$C_{1k} = \gamma_0 + \sum_{u=1}^{h} \gamma_{i_u} C_{i_u k} \phi_{jk} \tag{23}$$

We may refer to this set of equations as the **TMBR model**. Again, if this set of equations has rank $h+1$ then we may solve for the gamma coefficients and consequently calculate the individual terms of the equations. This will yield the apportionment we seek. Note that if we take $\phi_{jk} = 1$ then this set of equations reduces to the set of equations in (22).

Tracer Mass Balance (TMB) Model

This is a special case of the TMBR model and is obtained by partitioning the sources contributing a particular secondary aerosol species, (say species $i = 1$ with associated parent species designated as species $i^* = 2$), into two groups rather than $h+1$ groups. That is, we take $h = 1$ in the TMBR model. The two groups are: (i) A distinguished source labeled $j = 1$ with associated tracer species $i = i_1$, and (ii) All other sources. In this case, the TMBR model reduces to

$$C_{1k} = \beta_{0k} + \beta_{i_1 k} C_{i_1 k} \tag{24}$$

As before , if we assume that the beta coefficients are independent of the sampling period, then the TMB model equations further reduce to

$$C_{1k} = \beta_0 + \beta_{i_1} C_{i_1 k} \tag{25}$$

If the quantities C_{1k} and $C_{i_1 k}$ are known, and if the set of linear equations in (32) have rank 2 then we can solve for the unknown beta coefficients and consequently carry out the apportionment of species 1 by computing the individual terms of the above equations.

In certain instances it is known that the beta coefficients will differ significantly from one time period to another. In such cases it may be possible to determine, based on physical and chemical reasons, a function of the field measurements, the sampling period and the source, which we denote by ϕ_{1k}, such that it is more reasonable to assume the quantities $\beta_{i_1 k}/\phi_{1k}$ are constant for all sampling periods rather than the quantities $\beta_{i_1 k}$. In such cases we define $\gamma_{i_1} = \beta_{i_1 k}/\phi_{1k}$. For uniformity of notation we define γ_0 to be equal to β_0. This results in the system of linear equations

$$C_{1k} = \gamma_0 + \gamma_{i_1} C_{i_1 k} \phi_{1k} \tag{26}$$

We may refer to the above system of equations as the **TMB model**. Again, if this set of equations has rank 2, then we may solve for the gamma coefficients and consequently calculate the individual terms of the equations. This will yield the apportionment we seek.

A Special Case

The simplest versions of the TMBR model use $\phi_{uk} = 1$ for all time periods and source groups. However, if K_c or K_d are dependent on other variables such as solar radiation, concentration of key atmospheric chemicals and so forth, it may be possible to chose a form of ϕ_{uk} that will linearize the TMBR model.

In apportioning a secondary aerosol, the constant $\beta_{i_u jk}$ derived from the GMB model had the form

$$\beta_{i_u jk} = \frac{r^*_{1jk}}{r_{i_u jk}} \frac{c_{2jk}}{c_{i_u jk}} \tag{27}$$

with

$$r^*_{ijk} = \frac{K_c(i^*, j, k)}{K_c(i^*, j, k) + K_d(i^*, j, k) - K_d(i, j, k)} \times$$

$$\{exp(-K_d(i, j, k)t_{jk}) - exp(-[K_c(i^*, j, k) + K_d(i^*, j, k)]t_{jk})\} \tag{28}$$

and

$$r_{i_u jk} = exp(-(K_c(i_u, j, k) + K_d(i_u, j, k))t_{jk}) \tag{29}$$

If the species i_u does not convert and its deposition rate is the same as that of the secondary aerosol species i being apportioned, then

$$r_{i_ujk} = exp(-K_d(i,j,k)t_{jk}) \tag{30}$$

so that the ratio r^*_{1jk}/r_{i_ujk} reduces to $K_c(i^*,j,k)t_{jk}$ after using the approximation

$$exp(x) \approx 1 + x \text{ (when } x \text{ is sufficiently small).} \tag{31}$$

The full infinite series expansion for $exp(x)$ is given by

$$exp(x) = 1 + x + \frac{x^2}{2!} + \frac{x^3}{3!} + \cdots$$

and a first order approximation has been used in (31). It is possible to use higher order approximations of $exp(x)$ in these derivations but this is not pursued here.

An an example of the above approximation consider a case where $K_c(i^*,j,k)$ is proportional to RH_{uk} with proportionality constant B_{i^*j}. Then the ratio r^*_{1jk}/r_{i_ujk} is equal to $B_{i^*j}t_{jk}RH_{uk}$ which gives

$$\beta_{i_ujk} = B_{i^*j}t_{jk}RH_{uk}\frac{c_{2jk}}{c_{i_ujk}}. \tag{32}$$

Defining

$$\gamma_{i_uk} = \beta_{i_uk}/RH_{uk} \tag{33}$$

and assuming that γ_{i_uk} are constant for all sampling periods rather than the quantities β_{i_uk} suggests the use of RH_{uk} as a linear factor in the TMBR model equation (23).

The use of RH as a linearization parameter does not necessarily imply that the RH dependence of K_c is grounded in some basic chemical process. Rather, in the case of SO_2 to SO_4 oxidation, RH may be thought of as a surrogate variable depicting the amount of time that SO_2 spends in contact with clouds where oxidation is accelerated. Therefore, assuming $RH_{uk} = RH_k$, the TMBR model for the SO_2 - SO_4 system becomes

$$C_{SO_4k} = \gamma_0 + \sum_{u=1}^{h} \gamma_{i_u} C_{i_uk} RH_k \tag{34}$$

where:

$C_{SO_4 k}$	=	concentration of sulfate sulfur for time period k
$C_{i_u k}$	=	concentration of trace element i_u for time period k
γ_{i_u}	=	fractionation coefficient associated with trace element i_u
γ_0	=	background concentration of the species being apportioned, due to all sources not accounted for explicitly.
h	=	the number of source groups or types, each source group having a unique tracer.
RH_k	=	the relative humidity at the sampling site for time period k.

γ_0 and the γ_{i_u}'s can be estimated by various least square estimation techniques. Since both independent and dependent variables have error associated with them ODR is the method of choice.

Model Calculations and Uncertainties

We outline two approaches for the calculation of uncertainties associated with estimated contributions and fractional contributions of the species of interest (species 1, say) by the source of interest $S_T(j = 1)$ and the associated uncertainties. The first approach is computationally intensive as it involves computer simulation while the second approach is computationally simpler but relies on several approximations being sufficiently accurate.

First Approach. In Equation (23), the quantities C_{1k}, $C_{i_u k}$ and ϕ_{uk} are all observed with error. We shall denote the true values by C_{1k}, $C_{i_u k}$ and ϕ_{uk} and the observed values by the quantities $\tilde{C}_{1k}$, $\tilde{C}_{i_u k}$ and $\tilde{\phi}_{uk}$. We then assume that

$$\begin{aligned} \tilde{C}_{1k} &= C_{1k} + \epsilon_{C_{1k}} \\ \tilde{C}_{i_u k} &= C_{i_u k} + \epsilon_{C_{i_u k}} \\ \tilde{\phi}_{uk} &= \phi_{uk} + \epsilon_{\phi_{uk}} \end{aligned}$$

The quantity $\epsilon_{C_{1k}}$ is a random error with mean 0 and standard deviation $\sigma_{C_{1k}}$. The quantity $\epsilon_{C_{i_u k}}$ is a random error with mean 0 and standard deviation $\sigma_{C_{i_u k}}$. Likewise, the quantity $\epsilon_{\phi_{uk}}$ is a random error with mean 0 and standard deviation $\sigma_{\phi_{uk}}$. All random errors are assumed to be normal and mutually independent.

The unknown quantities γ_0, and $\gamma_{i_u k}$ for $u = 1, \cdots, h$ are estimated using the method of Orthogonal Distance Regression (ODR) with input data consisting of the measured values as well as the measurement uncertainties. Estimates of the true values C_{1k}, $C_{i_u k}$ and ϕ_{uk} are also obtained as output from ODR. The estimated values will be denoted with 'hats' over the corresponding true values. Then the estimated value of C^*_{1uk}, denoted by $\hat{C}^*_{1uk}$ is given by,

$$\hat{C}^*_{1uk} = \hat{\gamma}_{i_u} \hat{C}_{i_u k} \hat{\phi}_{uk}.$$

From this we obtain the estimated fractional contribution F_{uk} of species 1 by source group u for sampling period k as

$$F_{uk} = \frac{\hat{C}^*_{1uk}}{\hat{C}_{1k}}.$$

The estimated fractional contribution for the entire sampling period, by source group u, is denoted by F_u and is calculated as

$$F_u = \frac{\sum_{k=1}^{s} \hat{C}^*_{1uk}}{\sum_{k=1}^{s} \hat{C}_{1k}}.$$

To calculate the uncertainties to be associated with these estimates we may use the following procedure. We construct several (say, 100) synthetic data sets by perturbing the estimates of the true values C_{1k}, $C_{i_u k}$ and ϕ_{uk} using gaussian random deviates with mean zero and standard deviations equal to the respective measurement uncertainties. Each such synthetic data set is subjected to an ODR analysis to obtain estimates of contributions and fractional contributions of the various source groups to the receptor as explained above. This procedure results in a whole collection of estimates (say, 100) for the various quantities of interest. The root mean square error is then calculated for each quantity of interest using the collection of estimates obtained from perturbed synthetic data sets and using the initial estimates obtained from the actual data set as if they were the true values. This root mean square error associated with a given quantity of interest is used to quantify the uncertainty associated with that quantity. Recall that if θ represents the true value of a quantity and θ^*_q represents an estimate of θ obtained from the q^{th} synthetic data set, then the root mean square error is calculated by

$$\textit{Root Mean Square Error} = \sqrt{\frac{1}{Q}\sum_{q=1}^{Q}(\theta^*_q - \theta)^2}$$

Alternatively we may quantify the uncertainty associated with a given estimate using confidence intervals but we do not discuss that approach here.

Second Approach. In this section we discuss an approximate method of calculating the uncertainties associated with the model outputs. The concentrations C^*_{1uk} of species 1 (secondary species of interest) associated with each trace element i_u for each time period may be calculated by multiplying the measured values of $A_{i_u k} = C_{i_u k}\phi_{uk}$ for each trace element by the respective estimated regression coefficients as follows. ($\hat{\gamma}_0$ would just be the estimated intercept representing the estimated contribution from all sources not explicitly accounted for by any of the reference species used in the TMBR model.)

$$\hat{C}^{*}_{1uk} = \hat{\gamma}_{i_u} \times A_{i_u k} \tag{35}$$

The uncertainties for each of these concentrations $\hat{C}^{*}_{1uk}$ may be calculated by:

$$\sigma_{C^{*}_{1uk}} = \sqrt{A^2_{i_u k}\sigma^2_{\gamma_{i_u}} + \gamma^2_{i_u}\sigma^2_{A_{i_u k}} + \sigma^2_{\gamma_{i_u}}\sigma^2_{A_{i_u k}}} \tag{36}$$

The quantities $\sigma_{A_{i_u k}}$ are the uncertainties in the measured values $A_{i_u k}$ and is assumed to be known. In the special case discussed in the previous section, these uncertainties are part of the WHITEX data base. The quantities $\hat{\gamma}_{i_u}$ may be obtained as outputs from the regression packages that are used. Errors in $A_{i_u k}$ and the estimated regression coefficients have been assumed to be independent in the calculation of Equation (36).

The total calculated amount of species 1, C_{1k} for each time period is the sum of the C^{*}_{1uk}'s summed over all the reference aerosol species i_u and the intercept $\hat{\gamma}_0$.

$$C_{1k} = \hat{\gamma}_0 + \sum_{u=1}^{h} \hat{C}^{*}_{1uk} \tag{37}$$

The uncertainty associated with the total calculated concentration of species 1 for each time period is:

$$\sigma_{C_{1k}} = \sqrt{\sigma^2_{\gamma_0} + \sum_{u=1}^{h} \sigma^2_{C^{*}_{1uk}}} \tag{38}$$

assuming the covariance terms arising in the derivation are negligible.

The estimated fraction F_{uk} of species 1 from each source for any given time period is equal to the amount of species 1 associated with the trace element divided by the total calculated concentration of species 1:

$$F_{uk} = \frac{\hat{C}^{*}_{1uk}}{C_{1k}} \tag{39}$$

The uncertainty for each of these fractions is:

$$\sigma_{F_{uk}} = \sqrt{\frac{\sigma^2_{\hat{C}^{*}_{1uk}}}{C^2_{1k}} + \frac{\hat{C}^{*2}_{1uk}\sigma^2_{C_{1k}}}{C^4_{1k}}} \tag{40}$$

The mean fraction $\overline{F}_u$ of species 1 attributed to each source type is estimated by the mean species 1 concentration $\overline{C}_u$ for that source type divided by the mean total calculated concentration of species 1, $\overline{C}$, as follows:

$$\overline{F}_u = \frac{\overline{C}_u}{\overline{C}} \tag{41}$$

where

$$\overline{C}_u = \frac{1}{s}\sum_{k=1}^{s} \hat{C}^*_{1uk} \tag{42}$$

and

$$\overline{C} = \frac{1}{s}\sum_{k=1}^{s} C_{1k}. \tag{43}$$

The uncertainties for $\overline{C}_u$ and $\overline{C}$ are calculated by:

$$\sigma_{\overline{C}_u} = \frac{1}{K}\sqrt{\sum_{k=1}^{s} \sigma^2_{\hat{C}^*_{1uk}}} \tag{44}$$

and

$$\sigma_{\overline{C}} = \frac{1}{K}\sqrt{\sum_{k=1}^{s} \sigma^2_{C_{1k}}}. \tag{45}$$

The uncertainties associated with the mean fractions $\overline{F}_u$ are calculated by

$$\sigma_{\overline{F}_u} = \sqrt{\frac{\sigma^2_{\overline{C}_u}}{\overline{C}^2} + \frac{\overline{C}^2_k \sigma^2_{\overline{C}}}{\overline{C}^4}}. \tag{46}$$

The uncertainty formulas are all derived using propagation of error methods and assuming the covariances between various terms occurring in the derivation are negligible. These assumptions will not be true in practice and so the usefulness of the above approximations will depend upon how severely the assumptions used in the above derivations are violated.

Model Assumptions

The TMBR model assumptions are:

- The chemical species used as tracers in the model are assumed to be uniquely emitted by non-overlapping groups of sources. In particular none of the species other than the tracer associated with the source of interest can be emitted by another source unless there is an independent method such as CMB to partition the ambient species concentrations into components attributable to the various groups of sources.
- The composition of source emissions are constant over the period of ambient sampling.

- Deposition and conversion are constant from one sampling period to the next for each subgroup u.
- Measurement errors are random, uncorrelated, and normally distributed.

For the special case where k_c was assumed to be proportional to RH the additional assumptions are:

- Exponential forms of deposition and conversions can be represented by first order approximations.
- The RH at the receptor site is indicative of the amount of time that air parcels spend in contact with clouds and therefore can be used as an indicator of oxidation rate.

Potential Deviations from Assumptions

It is highly unlikely that deposition and conversion are constant in space and time and in many cases one can expect source profiles to change over the course of the study. These assumptions are implicit to the assumption that background and fractionation coefficients are time independent. Whether of not a linearization scheme is appropriate can be examined through goodness of fit tests of the proposed model and possible by direct experimental verification. The uniqueness of tracer species can be assessed by source testing and by releasing unique tracers from sources of interest.

Deviation from any of the assumptions will increase the calculated uncertainty in the final apportionments. The extent to which the inflation of uncertainty occurs will depend on how variable the regression coefficients are. Research into the effect of deviation from assumptions on apportionments is needed.

Model Inputs

The model requires the following quantities as inputs:

- The ambient concentrations of the aerosol species being apportioned.
- The ambient concentrations of the reference tracer species.
- Relative humidity at the receptor for each of the sampling periods, when $\phi_{uk} = RH_{uk}$ is used in the model rather than $\phi_{uk} = 1$.

- The uncertainties in the above quantities, when ODR is used to estimate the γ coefficients, rather than OLS.

Model Outputs

The model outputs include:

- Estimates of the actual amount of the contribution and the fractional contribution of the aerosol species of interest by the source or source type of interest to the receptor, along with the associated uncertainty estimates.
- Estimates of the average amount and the average fractional amount of the aerosol species of interest contributed by each source or source type of interest along with the associated uncertainty estimates.

Differential Mass Balance (DMB) Model

The DMB model is a receptor model combined with elements of a deterministic model. In this approach dispersion is accounted for by ratioing ambient trace material concentrations attributed to a source by known trace material release rates while deposition and conversion are explicitly calculated. The name "Differential Mass Balance" refers to the use of difference in trace material concentration to account for dispersion.

Model Equations

Suppose a particular source is of interest and we wish to determine the fractional contribution of some aerosol species to the receptor by that source. We shall designate the aerosol species of interest by the subscript i_1 and the source of interest by j. If species i is a secondary species then the corresponding parent species will be denoted by the subscript i^*. For example, if SO_4 is of interest, then i stands for SO_4 and i^* stands for SO_2. We are then interested in the quantity C_{ijk} for each of the sampling periods. We have, from Equation (2) that

$$C_{ijk} = c_{ijk} r_{ijk} d_{jk} + c_{i^*jk} r^*_{ijk} d_{jk} \,. \tag{47}$$

If i represents a primary species, then r^*_{ijk} is zero for all k. If i represents a secondary aerosol species that is not emitted as a primary aerosol, the quantity c_{ijk} is zero for all k. Therefore, the above equation simplifies to

$$C_{ijk} = c_{ijk} r_{ijk} d_{jk} \tag{48}$$

when i is a primary species and

$$C_{ijk} = c_{i^*jk} r^*_{ijk} d_{jk} \tag{49}$$

when i is a secondary species. A characteristic feature of DMB model applications is that the dispersion factor d_{jk} is determined based on field measurements. If a unique tracer is available for source j then d_{jk} may be calculated based on this unique tracer. It can also be calculated based on a reference aerosol species that may not be a unique tracer for source j by first calculating the amount of this reference species contributed to the receptor by the source of interest. Chemical mass balance model may be applied for this purpose. Other approaches are also possible.

The following discussion assumes that a unique tracer is available for source j of interest. This source will be referred to as S_t. The tracer material may be a naturally emitted primary aerosol species or may be introduced artificially. The aerosol species is denoted by the subscript i_0. Therefore Equation (48) becomes

$$C_{i_0jk} = c_{i_0jk} r_{i_0jk} d_{jk}. \tag{50}$$

Dividing the quantity C_{ijk} by the quantity C_{i_0jk} we get,

$$\frac{C_{ijk}}{C_{i_0jk}} = \frac{c_{ijk}}{c_{i_0jk}} \frac{r_{ijk}}{r_{i_0jk}} \tag{51}$$

when species i is a primary aerosol and

$$\frac{C_{ijk}}{C_{i_0jk}} = \frac{c_{i=jk}}{c_{i_0jk}} \frac{r^*_{ijk}}{r_{i_0jk}} \tag{52}$$

when species i is a secondary aerosol. It follows from this that

$$C_{ijk} = \frac{c_{ijk}}{c_{i_0jk}} \frac{r_{ijk}}{r_{i_0jk}} C_{i_0jk} \tag{53}$$

for primary aerosols i and

$$C_{ijk} = \frac{c_{i=jk}}{c_{i_0jk}} \frac{r^*_{ijk}}{r_{i_0jk}} C_{i_0jk} \tag{54}$$

for secondary aerosols.

Since aerosol component i_0 is a tracer for source j, the quantity C_{i_0jk} is the same as the quantity C_{i_0k} which is the ambient concentration of species i_0 at the receptor and can be measured. If furthermore the quantities $K_d(i,j,k)$, $K_c(i,j,k)$ are known when species i is primary, or, $K_c(i^*,j,k)$, $K_d(i^*,j,k)$ and $K_d(i,j,k)$

are known when species i is secondary, and if in addition, $K_d(i_0, j, k)$, $K_c(i_0, j, k)$, t_{jk} as well as the ratio $c_{i=jk}/c_{i_0jk}$ are known, then the contribution of the source of interest to the concentrations of the species of interest at the receptor can, in principal, be calculated.

If T represents a unique nonconverting, nondepositing tracer for source $j = 1$, then for a species that is directly emitted by source $j = 1$, Equation (53) for primary aerosols reduces to

$$C_{i1k} = \frac{c_{i1k}}{c_{T,1,k}} r_{i1k} C_{T,k}. \tag{55}$$

If the ratio $c_{i1k}/c_{T,1,k}$ is known, the form of r_{i1k} is

$$r_{i1k} = exp(-K_d(i, 1, k)t_{1k}).$$

For a species that is not directly emitted, but is a secondary species which is absent at the source, the equation for the DMB reduces to

$$C_{i1k} = \frac{c_{i^*1k}}{c_{T,1,k}} r^*_{i1k} C_{T,k}. \tag{56}$$

The ratio $c_{i^*1k}/c_{T,1,k}$ is assumed known and the form of r^*_{i1k} in this case is

$$r^*_{i1k} = \frac{K_c(i^*, 1, k)}{K_c(i^*, 1, k) + K_d(i^*, 1, k) - K_d(i, 1, k)} \times$$

$$\{exp(-K_d(i, 1, k)t_{1k}) - exp(-[K_c(i^*, 1, k) + K_d(i^*, 1, k)]t_{1k})\}$$

where

$K_c(i, 1, k)$ = conversion rate of species i from source 1 to its secondary form, during sampling period k.

$K_d(i, 1, k)$ = deposition rate of species i from source 1 during sampling period k.

Considering a specific example for SO_4 and SO_2 Equation (56) becomes

$$C_{SO_4,1,k} = \frac{c_{SO_2,1,k}}{c_{T,1,k}} r^*_{SO_4,1,k} C_{T,k} \tag{57}$$

and

$$C_{SO_2,1,k} = \frac{c_{SO_2,1,k}}{c_{T,1,k}} r_{SO_2,1,k} C_{T,k} \tag{58}$$

where

$$r^*_{SO_4,1,k} = \frac{K_c(SO_2, 1, k)}{K_c(SO_2, 1, k) + K_d(SO_2, 1, k) - K_d(SO_4, 1, k)} \times$$

$$\{exp(-K_d(SO_4,1,k)t_{1k}) - exp(-[K_c(SO_2,1,k) + K_d(SO_2,1,k)]t_{1k})\} \quad (59)$$

and

$$r_{SO_2,1,k} = exp(-(K_c(SO_2,1,k) + K_d(SO_2,1,k))t_{1k}). \quad (60)$$

From now on we shall use the notation $K_c = K_c(SO_2,1,k)$, $K_1 = K_d(SO_2,1,k)$ and $K_2 = K_d(SO_4,1,k)$. Furthermore, these parameters may be related to deposition velocities v_1 for SO_2 and v_2 for SO_4, and SO_2 oxidation rate K_c by the equations

$$K_1 = \frac{v_1}{H_m} \quad (61)$$

$$K_2 = \frac{v_2}{H_m} \quad (62)$$

where H_m is the mixing height. K_1, K_2, or K_c may be functions time of day, surface conditions, meteorological conditions, relative humidity and a number of other variables.

Model Calculations

Again consider the specific example of calculating the fractional contribution of SO_4 that is associated with a specific source emission of SO_2. The contribution of SO_4 by the source of interest ($j = 1$) to the receptor is calculated using Equation 57. The value of $c_{SO_2,1,k}/c_{T,1,k}$ is estimated from field measurements. The plume ages t_{1k} can be estimated from plume streakline analysis. $C_{T,1,k}$ are ambient concentrations of unique tracer, T, at the receptor and are assumed to be measured during the experiment. The values of K_c, K_1 and K_2 are unknown and may be estimated based on literature values of deposition velocity for SO_2 and particles and pseudo-first-order SO_2 oxidation rates. Alternatively, they may also be empirically derived from the measurements made during the experiment.

To judge if a particular combination of these parameters is consistent with the field measurements, the following procedure may be adopted. Using the chosen combination of values for these parameters we first calculate the SO_4 contributions $C_{SO_4,S_t,k}$ of source $j = 1$ for each sampling period. This procedure can be repeated for as many sources for which there are unique tracers or for as many sources for which the relative contribution of those sources to an ambient trace element concentration have been established. Relative contributions of sources to a specific tracer species could be established by CMB or deterministic approaches.

Therefore, the regression model

$$C_{SO_4,k} = \beta_0 + \beta_1 C^*_{SO_4,1,k} + \sum_{u=2}^{h} \beta_u C^*_{SO_4,i_u,k} + error \quad (63)$$

may be fitted and the adequacy of the fit judged by the resulting R^2 value and the closeness of the beta coefficients to one. $C^{*}_{SO_4,iu,k}$ refers to the total contribution of SO_4 by source group u to the receptor. If the chosen parameter combination results in a high R^2 value and beta values are not significantly different from one, then the chosen parameter values v_1, v_2, K_c may be judged as being consistent with observed data. The best possible value of R^2 obtained, by varying the values of v_1,v_2 and K_c over their entire range of values suggested in the literature, may be denoted by R^2_{opt}. The values $v_1 = v_{1,opt}$, $v_2 = v_{2,opt}$, and $K_c = K_{c,opt}$ which result in the best R^2 may be used to calculate the daily S_t contributions to SO_4 and SO_2 at the receptor. By calculating the ratio of the total S_t contribution over the entire sampling period to total ambient concentrations over the same period we can calculate the fractional SO_4 and SO_2 contributions by S_t during the experimental period.

Uncertainty Calculations

Uncertainties in the final results are primarily due to three sources.

- Uncertainties in t_jk.
- Uncertainties in the model parameters such as K_c, K_1 and K_2.
- Uncertainties in the measured values.
- Uncertainties in the extent to which the model assumptions are violated.

Uncertainties in the Model Parameters. The model parameters in question are K_c, K_1 and K_2 which are not known. Suppose a review of the literature suggests deposition velocities v_1 for SO_2 ranging from l_1 to u_1 cm/sec and v_2 for SO_4 ranging from l_2 to u_2 cm/sec. In addition suppose the sulfur dioxide oxidation rates varied from $K_c = l_c$ to $K_c = u_c$ percent per hour.

Clearly, not all combinations of values of v_1,v_2 and K_c are physically possible. To judge which combinations of these parameters are reasonable, the following procedure may be adopted. A grid of values for v_1,v_2 and K_c may be chosen by taking all possible combinations of these parameters resulting from

$v_1 = l_1$ to u_1 in increments of δ_1.

$v_2 = l_2$ to u_2 in increments of δ_2.

$K_c = l_c$ to u_c in increments of δ_c.

To decide whether a particular combination of values of v_1,v_2 and K_c are reasonable the regression model suggested by Equation 63 can be exercised and the adequacy of the fit may be judged by closeness of beta values to one and the resulting R^2. The best possible value of R^2 for β values close to one over the range of these parameters is denoted by R^2_{opt}. A value R^2_0 less than R^2_{opt} but close to it is chosen, based on subjective judgement, as a criterion value for judging the reasonableness of various combinations of the parameter values. Parameter combinations resulting in an R^2 equal to R^2_0 or greater may be considered reasonable. The set of all such parameter combinations will be denoted by the symbol $\mathcal{A}$. S_t contributions can be calculated for each of the parameter combinations in the set $\mathcal{A}$. This will result in a whole range of values for the daily S_t contributions and the overall average S_t contributions. The mean and the standard deviation for this range of values (as well as the minimum and the maximum values) may be calculated to assess the uncertainty in the estimated S_t contributions due to imprecise knowledge of the model parameters. The measured values of concentrations of species are assumed to be exact in these calculations.

Uncertainties in the Measured Values. To assess the effect of errors in measurements on the estimated S_t contributions to SO_4 and SO_2 at the receptor, the values of v_1,v_2 and K_c are fixed at their optimum values obtained as explained in the previous subsection. The measured values used in the calculations are: (1) The ambient T concentration, $C_{T,k}$, (2) The ambient SO_4 concentration $C_{SO_4,k}$, (3) The ambient SO_2 concentration $C_{SO_2,k}$, (4) Relative Humidity RH_k at the receptor, and (5) Transport time $t_{S_t,k}$ for the aerosol mixture from S_t to arrive at the receptor. Suppose each of these measurements have associated with them a standard deviation characterizing the uncertainty in the respective measurements. We generate a number of synthetic data sets (one hundred is sufficient for most purposes) on the computer by perturbing the measured values using random gaussian deviates with zero means and standard deviations associated with each of the measured values. For each synthetic data set thus generated, the daily S_t contribution to SO_4 and SO_2 at the receptor as well as the average contributions over the entire sampling period are calculated. The range of values thus obtained for each of these quantities gives an indication of the uncertainty that would be due to imprecise measurements alone. The results are reported in the form of means and standard deviations of each of the quantities of interest calculated from the synthetic data sets. Throughout this exercise, the model parameters, viz, the conversion and deposition parameters, are to be kept constant at their optimum values.

Uncertainties in the Extent to which the Model Assumptions are Violated. Assessment of the uncertainties in reported results arising from model

violations can be evaluated by conducting extensive sensitivity studies involving various perturbations in the model assumptions themselves.

Overall Uncertainties. Since the first two categories of uncertainties are expected to be "independent", the total uncertainty due to these two sources may be characterized by the effective total standard deviation

$$\sigma_{Total} = (\sigma_1^2 + \sigma_2^2)^{1/2}$$

where σ_1 and σ_2 are the standard deviations associated with the two categories of uncertainties respectively.

Model Assumptions

The DMB model assumes that the rates for deposition and conversion processes in the atmosphere are first-order and invariant in space and time. In particular, these rates are assumed to be constant at every point in space and time along the transport path. Deposition is dominated by dry deposition; precipitation scavenging is small by comparison. It is also assumed that the ratio of the emission rates for the species of interest (or its parent species) and the tracer is known.

Potential Deviations from Assumptions

Rates for deposition processes may not be first-order and invariant in space and time. For example, no dry deposition will occur until the plume has been mixed to the ground. Dry deposition velocities are known to vary depending on atmospheric stability, and the type of surface (vegetation or rock, dry or wet). Wet deposition during periods of intense precipitation may deposit more material than dry processes do. The ratio of the emission rates for the species of interest (or its parent species) and the tracer may not be known precisely.

Model Inputs

The inputs to the model are, as a function of time, the relative emission rates of SO_2 and tracer, ambient concentrations of tracer, primary and secondary aerosols, and plume ages, deposition velocities for particulates and reactive gases (v_d), mixing height (H_m), SO_2 pseudo-first-order oxidation rate.

Model Outputs

Ambient concentrations and fractions of total ambient concentrations of aerosols of interest associated with a given source.

Conclusion

A set of deterministic general mass balance (GMB) equations describing how primary and secondary aerosols and gases are transported and transformed as they pass through the atmosphere were formulated. From the GMB equations it is possible, with a variety of limiting assumptions, to derive the chemical mass balance, the differential mass balance equations, and the tracer mass balance regression model. Derivation of these receptor modeling approaches from a first principle model allows for an examination of model assumptions and deviations from assumptions. With assumptions identified it possible to make a better determination of how to incorporate measurement uncertainty and how to estimate model uncertainty associated with an imperfect knowledge of model parameters.

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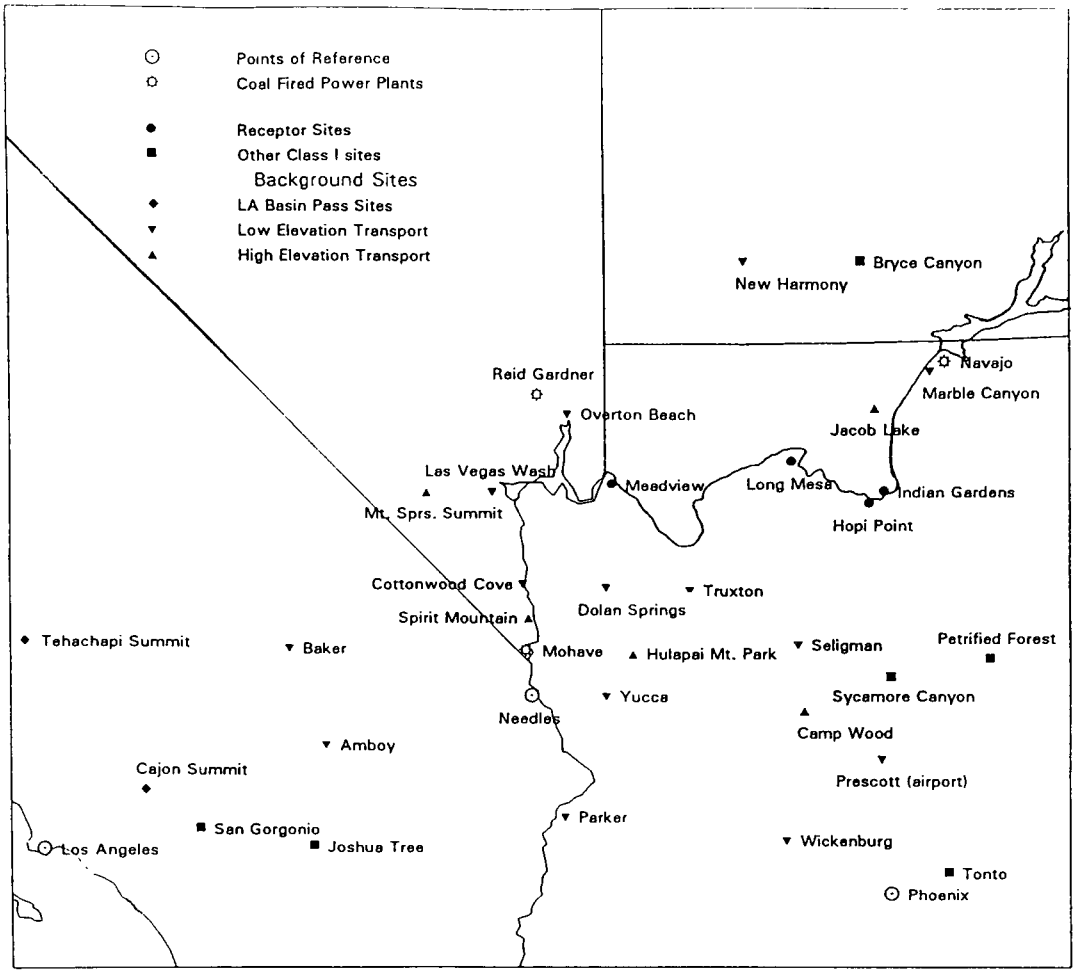
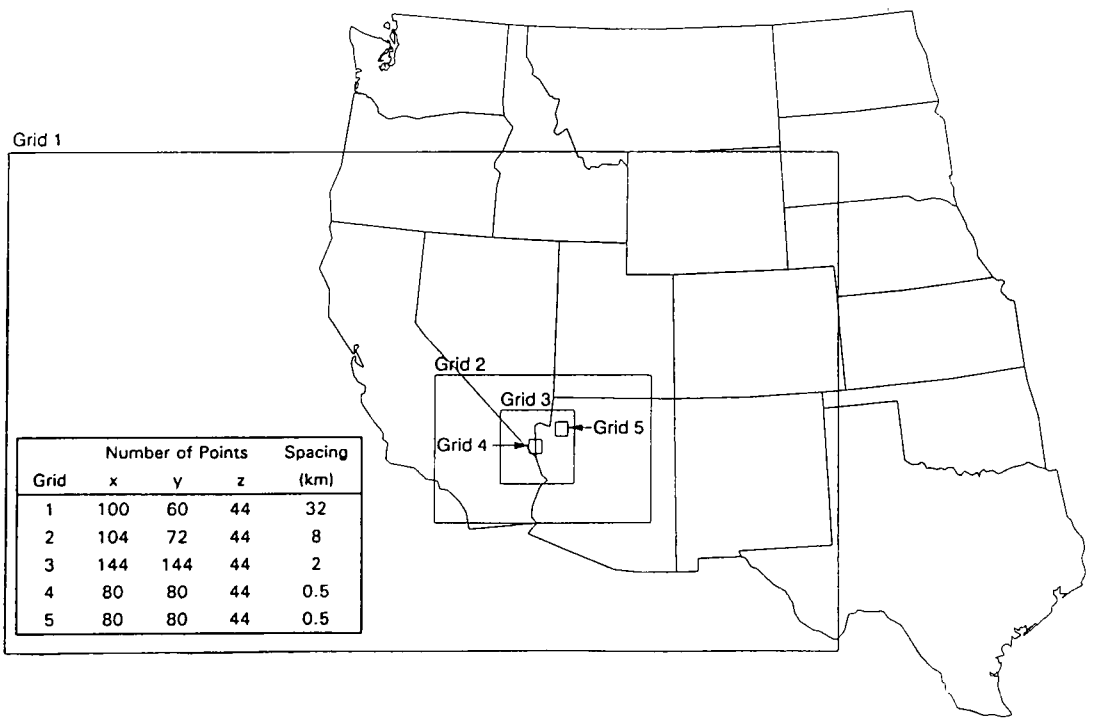
Project MOHAVE Summary

12/2/91

Study Component	Description of Study Component
Purpose and Objectives	Purpose: Respond to Congressional mandate for "Mohave Power Plant tracer study." Study Objectives: Estimate frequency and magnitude of any perceptible impact of Mohave Power Plant to visibility at Class I areas; Estimate impacts of other sources upon visibility in the southwest; Develop and evaluate tools for subsequent regional haze analyses.
Approach	Detailed intensive study periods nested within year-long study period. Results and conclusions to be based upon evaluation and reconciliation of multiple analysis approaches.
Schedule	Field Study: September 1991 November 1992 Winter Intensive: January 1992 (30 days) Summer Intensive: July August 1992 (50 days) Draft Report: July 1993 Final Report: December 1993
Tracer	Continuous stack release of perfluorocarbon tracer during intensives. Monitoring with 35 samplers at 31 sites. Release different tracers from southern California (Los Angeles Basin and San Joaquin Valley) during summer intensive.
Emissions	Continuous SO_x and NO_x stack monitoring during intensives. Detailed source profiling using daily samples during intensives.
Air Quality Monitoring	Full IMPROVE samplers at 10 sites, IMPROVE channel A + SO_2 at 21 sites during intensives (12 & 24 hour sampling). Sampling two days per week at 10 sites with IMPROVE samplers during non-intensives. DRUM sampling (8 size ranges, 6 hour resolution) at six sites during intensives. Sampling with medium volume particle samplers at three sites during intensives. Hydrogen peroxide sampling for a portion of summer intensive.
Optical Monitoring	Continuous monitoring entire study period. Nephelometers at all receptor sites, a transmissometer at Meadview, in addition to ones already at IMPROVE sites. Time-lapse photography at several sites.
Meteorological Monitoring	Continuous vertical wind profiling for 12 months at two sites using radar wind profilers. Two additional profilers during intensives. Surface meteorology at all wind profiler sites and receptor sites. Doppler sodar at two sites. RASS temperature profiling at two sites.
Deterministic Modeling	Deterministic meteorological modeling (wind, turbulence, moisture, etc.) every day for 12 month period. Calculation of influence functions. Detailed chemistry modeling (RADM, RPM) for selected cases. Monte Carlo transport modeling with linear chemistry every day for 12 month period.
Data Interpretation	Statistical study of historical sulfur concentrations and plant output. Spatial pattern (eigenvector) analysis. Hybrid receptor modeling utilizing artificial and endemic tracer data. Calculation of extinction budget. Reconciliation of modeling results. Source apportionment.
Quality Assurance	QA audit by independent reviewer covering all portions of the study.
Potential SCE Contributions	Upper air monitoring, particle monitoring (endemic tracers), chemical modeling, tracer release, data analysis, aircraft measurements, stack sampling, and data base management.



Monitoring Locations



Grid	Number of Points x	y	z	Spacing (km)
1	100	60	44	32
2	104	72	44	8
3	144	144	44	2
4	80	80	44	0.5
5	80	80	44	0.5

Meteorological Modeling Grids

For more information, contact Mark Green at (702) 798-2182.

December

Sunday	Monday	Tuesday	Wednesday	Thursday	Friday	Saturday
1	2	3	4 Begin returning tracer samplers to BNL	5	6	7
8	9	10 Finish returning tracer samplers to BNL	11	12	13	14 Begin analysis of tracer samples
15	16	17	18 Complete tracer analysis; Distribute background tracer data	19	20 Assess readiness;	21
22	23 Assess readiness	24	25 CHRISTMAS	26	27 Ship tracer	28
29	30	31				

January

Sunday	Monday	Tuesday	Wednesday	Thursday	Friday	Saturday
			1 NEW YEAR'S DAY	2	3 Ship tracer samplers	4
5	6 Begin deploying tracer and particulate samplers	7	8	9	10	11 Finish deploying samplers and begin tracer sampling
12	13	14 Begin tracer release; begin particulate sampling	15	16	17	18
19	20	21	22	23	24	25
26	27	28	29	30	31	

February

Sunday	Monday	Tuesday	Wednesday	Thursday	Friday	Saturday
						1
2	3	4	5	6	7	8
9	10	11	12	13 End tracer release	14	15
16 End particulate sampling	17	18	19	20 End intensive tracer sampling	21	22

Shaded days represent intensive sampling days. (prepared 12/17/91)

Schedule
Project MOHAVE Winter Intensive

DATE	ACTIVITY	ORGANIZATION
September - October	Begin year-round particulate monitoring at receptor and other Class I area sites; Begin year-round optical monitoring	UC-Davis; Air Resource Specialists
November - December	Install radar wind profilers/RASS	NOAA-Boulder
11/20 - 11/26	Deploy tracer samplers for background test	UC-Davis
12/4 - 12/9 or 12/10	Pickup tracer samplers, return to Brookhaven	UC-Davis
12/14-12/18	Analyze tracer samples	Brookhaven
12/18	Distribute background tracer data	Brookhaven
12/20, 12/23	**Assess readiness for field program; if OK, then the following schedule will hold. If major problems exist, re-evaluate study.**	**EPA, Brookhaven, NOAA-Idaho Falls, UC-Davis**
12/27	Ship tracer material to NOAA-Idaho Falls	Brookhaven
1/3	Ship tracer samplers to Lake Mead for winter intensive	Brookhaven
1/6 - 1/11	Deploy tracer and particulate samplers	UC-Davis
1/11 7am MST	Start tracer sampling	UC-Davis
1/14 7am MST	Start particulate sampling	UC-Davis
1/14 7am MST - 2/13 7am MST	Release tracer	NOAA-Idaho Falls
2/16 7am MST	Stop particulate sampling	UC-Davis
2/20 7am MST	Stop tracer sampling, except at Meadview and Hopi Point	UC-Davis

(schedule as of 12/17/91)