CHAPTER 1

INTRODUCTION

An intensive field monitoring program was undertaken during the summer of 1990 to study the contribution of emission sources to ambient particulate concentrations and regional haze at Federal Prevention of Significant Deterioration (PSD) Class I areas in the state of Washington. These areas included the national parks, managed by the National Park Service (NPS), namely Mount Rainier, North Cascades, and Olympic, and the wilderness areas managed by the U.S. Forest Service (USFS), namely Pasayten, Glacier Peak, Alpine Lakes, Goat Rocks, and Mount Adams. The acronym for the Pacific Northwest Regional Visibility Experiment Using Natural Tracers study is PREVENT.

This document presents a summary of the field measurement program, a summary of the data gathered during the field study, and an analysis of the collected data. Documents that have been prepared that describe the historical data from the area, program planning, field study implementation, and collected data are:

Air Resource Specialists, Inc., "PREVENT Preliminary Field Program Plan," April 1990.

------, "PREVENT Program Plan," September 1990.

------, "PREVENT Data Transmittal Report: Meteorological Data Collected From Operational Networks," December 1990.

------, "PREVENT Photographic Data Transmittal Report," February 1991.

------, "PREVENT Visibility and Meteorological Transmittal Data Report," February 1991.

Latimer, D.A., "Preliminary Conceptual Study Plan for PREVENT," 1990.

University of California at Davis, "Aerosol Data Report."

The detailed information contained in the above reports will not be repeated in this report but only summarized.

1.1 OBJECTIVES OF PREVENT

The primary objectives of PREVENT were:

1) To determine the spatial and temporal patterns of aerosol concentration, chemical composition, and particle size; regional emissions; and light extinction and observed visual effects.

2) To determine estimates of the light extinction budgets for the summer period for Mount Rainier and North Cascades National Parks.

3) To apportion (or attribute) the summertime haze observed in Federal Class I areas in Washington to the regional emissions from all sources in the Pacific Northwest and British Columbia.

4) To determine the contributions from natural and man-made sources.

1.2 STUDY AREA

Figure 1-1 is a map of the PREVENT study area. Federal Class I areas are shaded. Also shown are the total SO_2 emissions (tons/year) for each of the counties in the study area. Circles, whose areas are proportional to the total county SO_2 emissions, are also plotted on the map. Figure 1-2 shows the percentage of total 1988 SO_2 emission by county. The county with the largest SO_2 emission is Lewis, and the Centralia Power Plant constitutes most of the County's emissions. The Centralia Power Plant is located approximately 75 km due west of the southwestern corner of Mount Rainier National Park. The estimated annual SO_2 emissions from both of Centralia's stacks is 67,698 tons per year, or approximately 52% of the entire state of Washington's SO_2 emissions. Most of the remaining SO_2 emissions in the state are in King (12%), Skagit (7.6%), Pierce (7.5%), and Whatcom (7.5%) counties, all of which are in the prevailing upwind direction from Washington's Class I areas.

Although Mount St. Helens has been a rather large, natural SO_2 source in the past, (in 1987 its daily variable emissions have been estimated to range from 1,800 to 9,100 tons per year) current annual emissions are estimated to be less than 5,000 tons per year. Thus its emissions are expected to be about 7.0% of the Centralia plant's SO_2 emissions.

Slash and prescribed burning is a forest management tool used throughout the area. These burns are conducted by federal land management agencies, states, Indian tribes, and Canadian provinces. Wildfires can also occur throughout the region, depending on weather conditions. Smoke from these fires can cause visibility reduction in the study area.

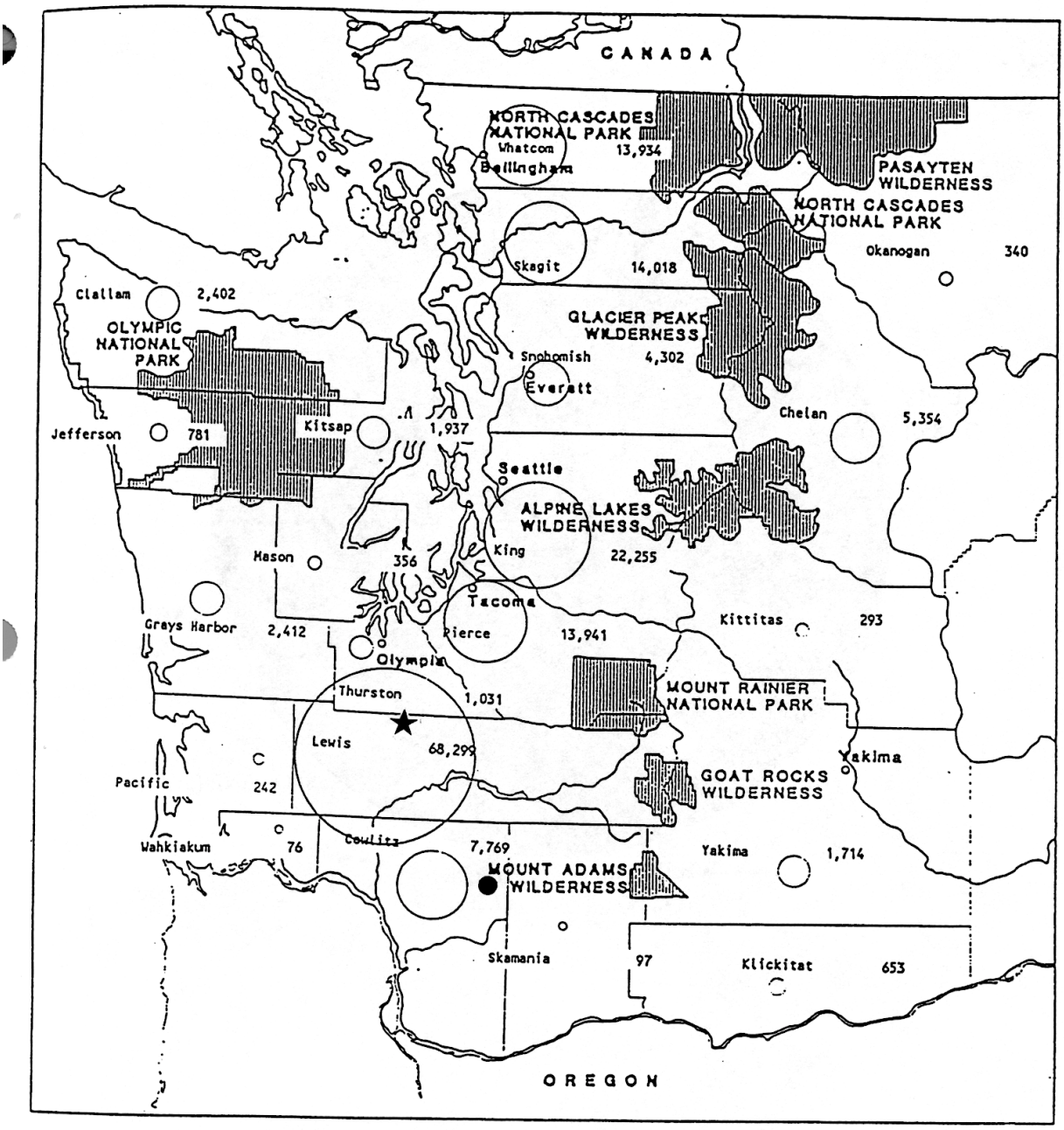

Figure 1-1. A map of the PREVENT study region. Federal Class I areas are shaded and the estimated 1988 SO_2 emissions (tons/year) are shown by county. Areas of circles shown are proportional to the 1988 SO_2 emissions estimated by the Washington Department of Ecology. ★ Centralia Power Plant ● Mount St. Helens

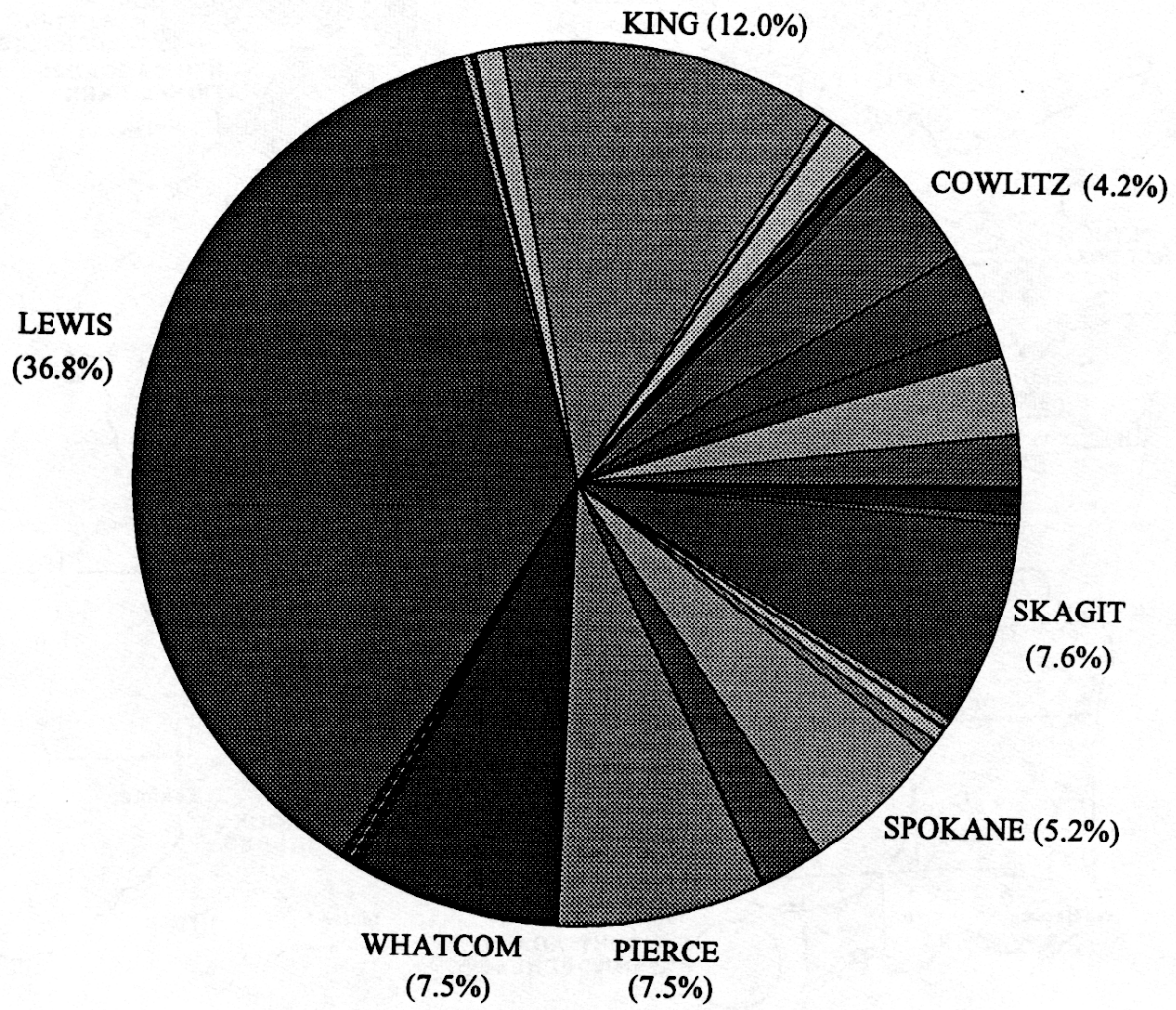

Figure 1-2. Percentage of total 1988 SO_2 emissions in the state of Washington by county. (Only the counties comprising 4.0% or more of the inventory are labeled; see Table 1-1 for emissions from all counties.)

1.3 REGULATORY BACKGROUND AND PRIOR STUDIES

In 1980, pursuant to the 1977 Clean Air Act, the Environmental Protection Agency (EPA) promulgated visibility protection regulations for national parks and wilderness areas that have been designated Federal Class I for Prevention of Significant Deterioration (PSD) of air quality. Among other things, these regulations required the states to develop programs to assure that reasonable progress was made toward the national visibility goal of remedying existing and preventing future visibility impairment.

Visibility data collected by the states of Washington and Oregon in the early 1980's clearly demonstrated that visibility is frequently impaired by man-made air pollution during the summer months when visitor use of Class I (and other) areas is at its peak. These studies indicated that regional haze constituted 70% of the impairment during daylight hours and that long-range transport of smoke from forest slash burning, often across state borders, was an important issue.

The Oregon Department of Environmental Quality (ODEQ) studied visibility impairment in Oregon's Class I areas.[1] Class I areas were found to be frequently impaired by uniform haze and, to a lesser extent, ground-based layered haze, with plume blight from coherent plumes very uncommon. During the 1982-84 period, one-third of the uniform or layered haze impacts were due to burning, and one-third from regional haze. Field and slash burning were found to be the principal contributing sources of man-made haze: 50% of the fine particle mass at Mount Hood was associated with slash burning while 20% originated from field burning. In the central Cascades, slash burning was estimated to contribute approximately 30% of the fine particle mass, while field burning contributed 25% to 30%. Interstate transport from Washington slash burning was found up to 25% of the time.

The Washington Department of Ecology (WA-DOE) has studied visibility since 1981.[2,3,4] Direct plume blight impacts were observed an average of 35 times a year during the summer months in or near western Washington's Class I areas. Simultaneous concentration maxima indicated that a regional haze was also common. A dominant fraction of the particle mass was carbon, probably associated with wood burning, while sulfate was the second most important species. Three tracer ratios were identified as having promise for future studies: K/Fe for forest burning, Cu/Pb for copper smelters, and Mn/V for coal-fired power plants.[4]

In order to obtain additional data regarding the regional haze issue in the Pacific Northwest, the states of Washington, Oregon, and Idaho, in cooperation with EPA Region X, the NPS and the USFS, embarked in 1984 on a major study, the Pacific Northwest Aerosol Mass Apportionment Study (PANORAMAS). This study was designed to:

1) define the geographical extent of regional haze in the region;
2) determine the frequency of haze episodes;
3) measure fine particle mass and chemical composition;
4) identify the sources of fine particle emissions;
5) conduct transport and trajectory analyses; and most importantly,
6) develop source-receptor relationships necessary to quantify source contributions to regional haze.

Monitoring of fine particle mass, light scattering coefficient (b_{scat}), and standard visual range (SVR) was carried out between May and mid-November 1984. The dominant components of the fine particle mass that caused the regional haze were (in order of importance): organic carbon, sulfate, and the crustal elements (Si, Al, Fe, Ca, Ti). Field, slash, and wildfire burning was found to be the major source of summertime pollution in the Pacific Northwest, accounting for approximately 62% of the fine particle mass and 40% to 50% of the light extinction. Sources of sulfur dioxide (SO_2) and primary sulfate responsible for the sulfate aerosol were the second most dominant cause of summertime haze, contributing 15% of the fine particle mass. Impacts were found to be episodic, resulting from local, subregional, and regional sources. One of the major suggestions to come from PANORAMAS was that "source apportionment to specific sources within the identified source categories" be carried out.[5]

In recent years, there has been a documented improvement in visibility both in Oregon and Washington. Core[6] reported that during the summer of 1988 substantial visibility impairment (defined as $b_{ext} > 80$ Mm^{-1}) was only 30% as frequent as in the 1982-1984 period. This improvement was attributed largely to the total ban on slash burning during the July 4 through Labor Day period, which was first fully implemented in 1987. Van Haren[7] has also reported a significant reduction in visibility impairment during the past five years at South Mountain and Dog Mountain. He reported that burning in western Washington has decreased significantly during the past decade. In the State of Washington, a weekend ban on burning during the period from July 4 to Labor Day was first implemented in 1983.

1.4 EMISSIONS IN THE STUDY AREA

Table 1-1 summarizes the emissions by county of total and fine particulates, sulfur dioxide (SO_2), nitrogen oxides (NO_x), and volatile organic compounds (VOC) estimated by the Washington Department of Ecology.[8] Table 1-2 summarizes state emissions of total and fine particulates released from agricultural and forest burning in 1984 for the PANORAMAS states.

Figure 1-3 shows the significant reduction in prescribed fire tonnages that have occurred during the past decade. From 1984 (the year of PANORAMAS) to 1988, total prescribed fire tonnages in western Washington have decreased 40% (from 769,000 tons to 462,000 tons). A comparable reduction in regional SO_2 emissions also took place during this time period, with the shutdown in March 1985 of the ASARCO Tacoma smelter, which was estimated to emit 143,000 tons/year of SO_2.[9] One of the objectives of PREVENT was to study the effect of these significant changes in regional emissions from both burning and industrial sources of SO_2 that have occurred since 1984 when PANORAMAS was conducted.

Table 1-1. Emission inventory for chemical species (particulate, fine particulate, SO_2, NO_x, and VOC) relevant for visibility for the counties in the state of Washington. The data is from the Washington Department of Ecology (1990). Point source data represents 1988, and area source data represents early 1980's. Units are tons/year.

County	Total Particulate			Fine Particulate			SO_2			NO_x			VOC		
	Area	Point	Total	Area	Point	Total	Area	Point	Total	Area	Point	Total	Area	Point	Total
Adams	1451	294	1745	877	0	877	176	0	176	1731	0	1731	1901	0	1901
Asotin	987	21	1008	687	10	697	88	0	88	692	1	693	1329	2	1331
Benton	4649	756	5405	3886	176	4062	1428	2018	3446	6093	2012	8105	9905	63	9968
Chelan	2268	2460	4728	1765	1152	2917	487	4867	5354	3348	235	3583	5150	221	5371
Clallam	2959	655	3614	2340	283	2623	866	1536	2402	4128	800	4928	5263	602	5865
Clark	6262	2351	8613	5611	1051	6662	1807	3156	4963	9323	2355	11678	12663	1154	13817
Columbia Heights	417	0	417	286	0	286	42	0	42	444	0	444	618	0	618
Cowlitz	4137	4715	8852	3522	2223	5745	1145	6624	7769	6903	6684	13587	9438	3130	12568
Douglas	1293	336	1629	858	292	1150	134	912	1046	1164	274	1438	1700	615	2315
Ferry	559	152	711	285	54	339	66	3	69	449	50	499	826	25	851
Franklin	1765	141	1906	1329	69	1398	283	0	283	2704	0	2704	3746	1037	4783
Garfield	738	50	788	450	0	450	33	0	33	409	0	409	1021	0	1021
Grant	3011	82	3093	2228	0	2228	434	2	436	4528	1	4529	6284	158	6442
Grays Harbor	4004	1902	5906	3221	701	3922	1464	948	2412	6926	600	7526	8106	616	8722
Island	1278	90	1368	1043	0	1043	203	8	211	1669	42	1711	3007	27	3034
Jefferson	1524	577	2101	1131	396	1527	402	379	781	1648	411	2059	3158	144	3302
King	42111	2611	44722	0	1144	1144	20930	1325	22255	53633	3454	57087	80584	7385	87969
Kitsap	4120	167	4287	0	59	59	1474	463	1937	4586	222	4808	9206	507	9713
Kittitas	2302	262	2564	1748	97	1845	292	1	293	3382	84	3466	4864	57	4921
Klickitat	1404	1203	2607	885	566	1451	143	510	653	1421	459	1880	1817	387	2204
Lewis	4496	4510	9006	3643	3544	7187	601	67698	68299	5300	32091	37391	7632	557	8189
Lincoln	1522	3	1525	480	0	480	120	0	120	1379	0	1379	1815	0	1815
Mason	1435	206	1641	1088	48	1136	256	100	356	1902	113	2015	3783	39	3822
Okanogan	7325	530	7855	5102	139	5241	335	5	340	3371	75	3446	8088	96	8184
Pacific	1050	218	1268	781	148	929	242	0	242	1562	67	1629	2615	27	2642
Pend Oreille	481	275	756	322	95	417	62	241	303	591	254	845	937	12	949
Pierce	15485	3563	19048	0	1830	1830	9941	4000	13941	17943	2681	20624	41275	4392	45667
San Juan	120	54	174	28	11	39	46	2	48	191	2	193	983	2	985
Skagit	3089	1952	5041	2526	121	2647	843	13175	14018	5130	3725	8855	7141	5458	12599
Skamania	2169	275	2444	1599	32	1631	91	6	97	973	58	1031	2536	33	2569
Snohomish	10809	1516	12325	0	744	744	2088	2214	4302	11435	960	12395	26933	2021	28954
Spokane	17312	2642	19954	14391	1221	15612	2597	7020	9617	17333	314	17647	32163	1701	33864
Stevens	1263	1139	2402	769	504	1300	231	868	1099	1827	1469	3296	2290	585	2875
Thurston	4883	138	5021	4276	42	4318	1022	9	1031	7101	19	7120	10992	407	11399
Wahkiakum	334	55	389	215	23	238	76	0	76	641	0	641	727	0	727
Walla Walla	2338	2966	5304	1589	1810	3399	299	350	649	2370	1169	3539	4349	266	4615
Whatcom	3439	2471	5910	2887	1024	3911	1660	12274	13934	6424	4034	10458	9240	7842	17082
Whitman	2404	824	3228	1547	159	1706	260	425	685	2536	647	3183	3176	7	3183
Yakima	10508	6222	16730	7403	540	7943	1606	108	1714	10943	298	11241	16068	399	16467
Totals	177701	48384	226085	80825	20308	101133	54273	131247	185520	214133	65660	279793	353329	39974	393303

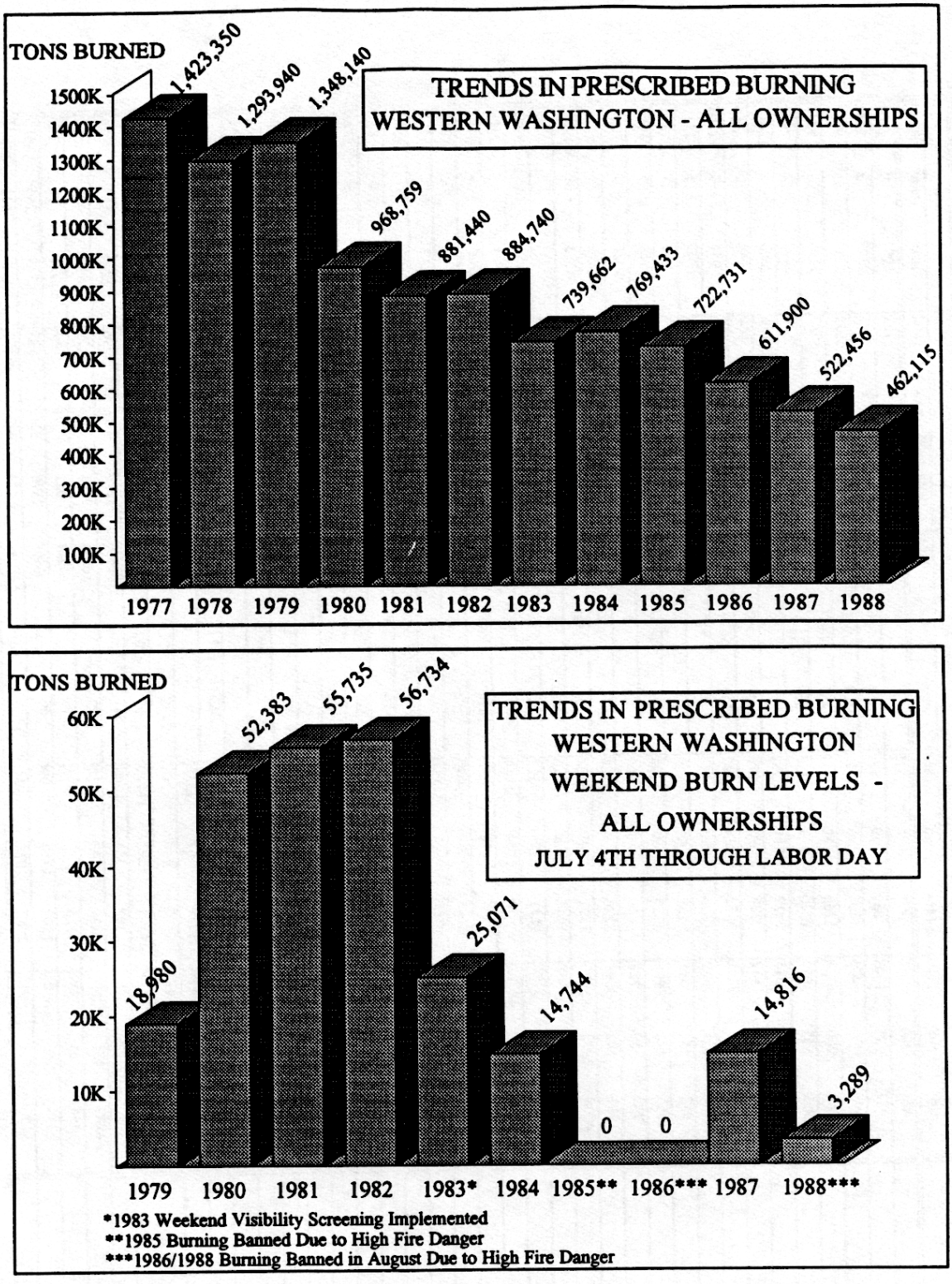

Figure 1-3. Trends in prescribed burning in western Washington, 1977-1988. (source: Van Haren[7])

Table 1-2. Agricultural and forest burning emission inventory summary for the Pacific Northwest states. Units are tons/year. (Source: Beck & Associates[5])

	Oregon		Washington		Idaho	
Fire Type	TSP	Fine*	TSP	Fine	TSP	Fine
Wildfires	12,182	9,746	7,330	5,864	42,727	34,182
Slash Burning	101,936	81,549	87,459	69,967	27,273	21,818
Field Burning	20,366	16,293	926	741	5,105	4,084
Totals	134,484	107,588	95715	76,572	75,105	60,084

*Fine particle fraction less than 2.5μm; estimated here as 80% of the total suspended particulate (TSP) emissions.

1.5 DATA ANALYSIS AND MODELING PROTOCOLS

In this section, the various data analyses and modeling techniques that were used on the PREVENT data set to meet the defined objectives are briefly described.

Apportionment of emissions to visibility, using receptor techniques, requires an underlying model. Because any model is an approximation of the underlying basic processes governing deposition, transport, and transformation, it is inherently an imperfect replica of reality. Therefore, to the degree possible, important assumptions should be explicitly listed, and when possible, the assumptions should be tested as to their effect on modeling results.

Visibility is more than specifying any one optical variable. The "seeing" of a landscape feature is dependent on the characteristics of that landscape as well as the optical properties of the atmosphere. The optical properties of the atmosphere are dependent on path radiance and atmospheric transmittance, which in turn are dependent on the extinction coefficient, volume scattering function, and illumination. The two variables that conceptually can be apportioned to emissions are the volume scattering function and extinction coefficient. However, because of the complexity associated with apportionment of the volume scattering function, all researchers to date have only attempted to apportion extinction, and from extinction, estimated path radiance using nominal volume scattering functions.

Apportionment of extinction to emissions is at least a two-step process: first, extinction must be apportioned to aerosols, and second, aerosols must be apportioned to sources.

The apportionment of extinction to aerosol species in and of itself is not an unambiguous process.[10] Whereas the sum of mass associated with individual species should equal gravimetric mass, the sum of scattering by individual species need not equal measured scattering. Because light scattered by a particle is dependent upon the size of the particle, it becomes almost nonsensical to talk about scattering by individual species when those species are mixed together to form a single

particle. Scattering by a mixture of particles is additive only when the index of refraction of the mixture is not a function of composition or size, and its density is independent of volume.[11] Nevertheless, it has become traditional to construct extinction budgets showing the extinction associated with each species.[12] In many, if not most studies, the sum of aerosol extinctions approximate measured extinction, and consequently assumptions may not have been grossly violated. The appropriateness of the apportionment scheme can only be judged within the context of whether the model is physically "reasonable," and whether independent apportionment of mass to extinction is consistent with measurements of scattering and absorption.

Further apportionment of extinction to an aerosol species from a specific source requires at least one further important assumption. The aerosol species attributed to a specific source has the same micro-physical-chemical characteristics as all other aerosol species of that type. For instance, the particle size distribution of ammonium sulfate attributed to source A is the same as all other sulfates found in the atmosphere. It is conceivable that removal of a fraction of "optically active" sulfur could disproportionally reduce sulfate scattering.

1.5.1 Light Extinction Budget

A light extinction budget is a breakdown of the total light extinction coefficient (b_{ext}) into the contributions from individual light scattering and absorbing gases and particles. The light extinction budget is determined from collocated particle and optical monitoring devices.

The light extinction budget can be determined solely by empirical means, using multiple regression analysis to determine the model that best fits the data. This technique is the most commonly used. The regression yields values of a_o and b_i in the following equation:

$$b_{ext} = a_o + \sum b_i C_i \qquad (1\text{-}1)$$

where a_o is the intercept (the light extinction without aerosol species i), b_i is the light extinction efficiency of species i, and C_i is the ambient concentration of species i.

However, the hygroscopic properties of sulfate, nitrate and organic aerosols must be included to account for liquid water associated with these species at higher humidities. With the purely empirical technique, the b_i must be adjusted, based on relative humidity, for example:

$$b_i(RH) = b_i(RH = 0)/(1 - RH)^a \qquad (1\text{-}2)$$

where RH is humidity expressed as a fraction (percent/100%) and the exponent a is commonly taken to be one.

In some cases, a more accurate light extinction budget can be estimated using theoretical fundamentals or by combining statistical and theoretical approaches (semi-empirical). For example, thermodynamic equilibrium considerations can be used to estimate the amount of liquid water associated with a given hygroscopic species as a function of relative humidity. However, sometimes this approach is quite uncertain because one may not know whether the aerosol is internally or externally mixed and to what extent hysteresis effects are occurring. If particle size information is available, Mie scattering calculations can be used to derive theoretical values for b_i.

For PREVENT, in order to determine the light extinction budget from collocated aerosol and optical measurements, a combination of empirical (statistical), semi-empirical, and theoretical approaches were used in parallel and intercompared.

1.5.2 Source Attribution Techniques and Models

Most visibility impairment is associated with secondary aerosols such as sulfates, nitrates, and secondary organics. Therefore, any visibility source apportionment scheme must address secondary as well as primary particles. Traditionally, the chemical mass balance (CMB) formalism has been used to apportion primary particles, and a number of review articles have addressed the associated assumptions.[13-15] It relies on known physical and chemical characteristic aerosols, such as ratios of tracer species, natural or man-made, at the receptors and sources to attribute aerosols to single sources or source types. Important assumptions associated with CMB are: composition of source emissions are known and constant; aerosol components do not react with each other (implicitly this means that the ratios of various tracer species associated with a source remain constant over time); the number of sources is less than the number of tracer species; and compositions of sources are linearly independent of each other. If any of these assumptions are violated, the apportionment is degraded.[16-18]

CMB can also be used to estimate a source contribution to secondary aerosols if additional assumptions are imposed: all primary gases are converted to the secondary aerosol of interest; the relative deposition of primary and secondary species is the same for all sources; the ratios of secondary aerosols to trace element species are equilibrated at constant values; the equilibrated ratios are in proportion to the relative emission rates of the primary gas; the net effect of these assumptions is to apportion the secondary aerosol in direct proportion to the apportioned primary mass; and the relative emission strength of each source. For instance, in a given sampling period, if 50% of the primary mass is associated with source A and source A emits 80% of all SO_2 emissions, then source A is estimated to contribute 40% of the ambient sulfate (0.5*0.8=0.4).

CMB modeling apportions aerosol species on a sampling-period-by-sampling-period basis. However, if the data set contains an adequate number of samples, regressional techniques, along with less restrictive assumptions, can be used to estimate apportionment of secondary species.[19-21] In a regressional approach, the secondary species is the dependent variable while the independent variables are tracers that are unique to a single source or group of sources. Assumptions associated with this approach are: the chemical species used as tracers are assumed to be uniquely emitted by non-overlapping 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, but they don't have to be the same across all sources; and measurement errors are random, uncorrelated, and normally distributed. The derivation of the CMB and regressional formalism from first principles can be found in Appendix 1. Major assumptions are listed and methods of linearizing the inherently nonlinear models are discussed. The regressional models are referred to as tracer mass balance regression (TMBR) models.

It is highly unlikely that the regression coefficients are constant for all sampling periods. This will inflate the uncertainty in the final apportionments, but the extent to which the inflation

occurs will depend on how variable the coefficients are. Whitmore *et al.*[22] explores the effects of source emissions being collinear with each other; the effect of variability of deposition and conversion; and meteorological factors on apportionment of secondary aerosols to a single source. Under worst case assumptions, the relative error was about a factor of two, while the average relative error, for variation in the input variables chosen was less than one.

If emitted tracer species are not unique to a given source or source type, then CMB can be used to partition the ambient concentrations into components attributable to the various source groups.

Hybrid models that combine elements of deterministic and receptor oriented models can be employed.[19,23-25] In this approach, dispersion is accounted for by rationing ambient trace material concentrations attributed to a source by known trace material release rates, while deposition and conversion are explicitly calculated. Assumptions associated with this approach are: the chemical species used as tracers are assumed to be uniquely emitted by non-overlapping groups of sources; the composition of source emissions are constant over the period of ambient sampling; and deposition and conversion rates are usually assumed to be first order and invariant in space and time. However, since transport time is explicitly taken into account, deposition and conversion can vary from one sampling period to the next.

Rates for deposition and conversion probably are not first order and invariant in space and time. For example, dry deposition will not occur until the plume has been mixed to the ground. Dry and wet deposition velocities are known to vary depending on atmospheric stability and on surface type. Conversion processes are dependent on other chemical species which vary in space and time. To date a systematic examination of the effect of assumptions associated with hybrid modeling has not been reported.

The objective of source attribution is to determine the fraction of each light-scattering or absorbing species contributed by a given source or source category. In PREVENT a variety of attribution techniques and models are used in parallel to:

- supplement each other;
- allow the strengths of one technique to offset the weaknesses and limitations of others; and
- reduce the overall uncertainties of the analysis.

For example, it has been recommended that both source (dispersion or deterministic) models and receptor models be used to expand the capabilities of each.[26] Elements of both receptor and deterministic models have been combined by Stevens and his colleagues at the EPA and by Malm and his colleagues at the NPS.[27] A review of the general mass balance equation and the derivation, along with associated assumptions, of CMB, TMBR, and hybrid models are presented in Appendix 1.

The next sections briefly discuss the specific analyses used in the PREVENT report to gain insight into the relationships between sources and visibility reducing aerosols.

1.5.2.1 Spatial and Temporal Analysis

Although not truly a quantitative technique, spatial and temporal trend analysis can be used to determine whether it is likely that a local source or a combination of regional influences is dominating a given impact. If concentrations are relatively homogeneous throughout a region and maxima occur at roughly the same time, one might expect distant source contributions. However, if concentrations are quite nonuniform and maxima occur at different times, it would be difficult to postulate that long-range transport is the key contributor rather than local sources.

1.5.2.2 Empirical Orthogonal Function (EOF) Analysis

This is a refinement of spatial and temporal trend analysis. EOF is a method of simplifying enormous quantities of spatial and temporal trends into a few simple patterns that are relatively easy to interpret. EOF analysis is a decomposition of a single data matrix (time x space) of ambient concentrations into two matrices: one which is independent of time (EOF by site); and one which is independent of space (time by EOF). Generally only a few EOFs are needed to explain most of the variance in concentrations in time and space, and EOF patterns can usually be interpreted in terms of known geographic distributions of emission sources.

1.5.2.3 Chemical Mass Balance (CMB) Modeling

CMB consists of a least-squares solution to a set of linear equations which expresses the ambient concentration of a given species at a given receptor as a linear sum of products of source profile species and source contributions. The source profiles are the fractional amount of each species in the emissions from each source type. Since different source categories can have quite different profiles, one can distinguish the contributions of individual species. Traditionally, CMB has been applied solely to conservative species (i.e., those that do not form in the atmosphere); however, there has been recent work that has attempted to combine elements of deterministic models and CMB to assess the contributions of source categories to secondary sulfate.

1.5.2.4 CMB Regression Analysis

Tracer mass balance regression (TMBR) 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. In this report CMB is used to apportion trace elements to sources and the apportioned trace elements, or relative source strengths, are then used in a TMBR analysis to make source apportionment estimates. This approach will be referred to as CMB regression analysis.

1.5.2.5 Trajectory Analysis

Wind field models are used to develop forward trajectories from sources and backward trajectories from receptor sites. These trajectories are used to examine validity of CMB and TMBR results, and are statistically analyzed to identify source areas associated with various tracers of opportunity and visibility reducing aerosols.

1.6 DATA REQUIREMENTS FOR ANALYSIS PROTOCOLS

The specific data needs for the analysis techniques described in the previous section dictated the requirements for ambient measurements during PREVENT and the collection of data from other networks. Table 1-3 presents the major measurement and data collection categories employed for PREVENT, and which analyses require the given data as input or as useful corroboration. Figure 1-4 presents a flow diagram of the interrelationships between ambient measurements and visibility cause-effect conditions that were studied. These categories are:

1) ambient concentrations ($SO_4^{=}$, NO_3^{-}, organic and elemental carbon, total fine and coarse mass, trace metals, SO_2, NO_x, HNO_3, O_3, etc.);

2) atmospheric optical parameters (b_{ext}, b_{scat}, b_{abs});

3) meteorology (upper air winds, temperature structure, humidity, clouds, precipitation); and

4) emissions (power plant load, hourly emissions, source profiles, including natural and artificial tracers, regional emission inventory).

Table 1-3. Measurement and data collection required for PREVENT as input for the various proposed analysis techniques for source attribution and light extinction budgets.

Measurements/Data	Spatial/Temporal Trends Emp. Orth. Func. (EOF)	Chemical Mass Balance (CMB)	Tracer Mass Balance (TMBR)	Deterministic Models Based On Windfields	Light Extinction Budget
AMBIENT CONCENTRATIONS					
Dense network of 24-hour avg. SO_4 concentrations	Essential	Not needed but useful	Not necessary	Useful	Not needed
12- or 24-hour avg. concentrations of all key gases, aerosols, and tracers (natural and artificial) at 3 or 4 receptors (e.g., 3 parks).	Not needed	Essential	Essential	Not necessary,but useful	Essential
Size distribution of ambient aerosol	Not needed	Not needed	Not needed	Not necessary, but useful for determining whether secondary sulfate is formed from gas- or aqueous-phase processes	Useful to calculate light scattering efficiencies
Concentrations of key oxidants (e.g., H_2O_2, O_3, and catalysts)	Not needed	Not needed	Not needed	Essential	Not needed
ATMOSPHERIC OPTICAL PARAMETERS					
12- or 24-hour avg. b_{ext}, b_{abs}, and b_{scat}	Not needed	Not needed	Not needed	Useful	Essential
Color slides and/or time-lapse movies to record key vistas and appearance of haze and plumes and cloud and fog conditions	Not needed	Not needed	Not needed	Useful to characterize whether aqueous-phase chemistry is occurring	Useful to identify periods of natural visibility impairment
METEOROLOGICAL PARAMETERS					
Upper-air (at plume height and throughout mixed layer) winds, temperature, and *RH* at sufficient density to characterize complex terrain effects and temporal variability	Not necessary, but useful to interpret spatial and temporal patterns	Not necessary	Not necessary, but useful	Essential	Not necessary,but helpful in interpreting sources of components of haze
Precipitation amount	Not needed	Not needed	Not needed	Needed to quantify wet deposition	Useful to identify periods of natural visibility impairment

Table 1-3. continued.

Measurements/Data	Spatial/Temporal Trends Emp. Orth. Func. (EOF)	Chemical Mass Balance (CMB)	Tracer Mass Balance (TMBR)	Deterministic Models Based on Windfields	Light Extinction Budget
EMISSIONS					
Hourly emissions of SO_2, NO_x, and fly ash at the Centralia Power Plant. Also boiler load and stack parameters.	Not needed	Not needed	Not needed	Essential	Not needed
Aircraft measurement of near-stack plume for Centralia and other major source categories.	Not needed	Desirable, but existing profiles could be used	Desirable, but existing profiles could be used	Not needed	Not needed
Emission inventory for all sources of SO_2, NO_x, VOC, and particulate, including fire episodes.	Not necessary, but useful for interpretation	Not needed	Not needed	Essential	Not needed
AIRCRAFT MEASUREMENTS OF PLUME CHEMISTRY					
Aircraft measurements of plume constituents to determine oxidation rates.	Not needed	Not needed	Not needed	Essential	Not needed
Aircraft measurements of cloud water chemistry to determine concentrations of key species (e.g., pH, H_2O_2, and O_3).	Not needed	Not needed	Not needed	Essential	Not needed

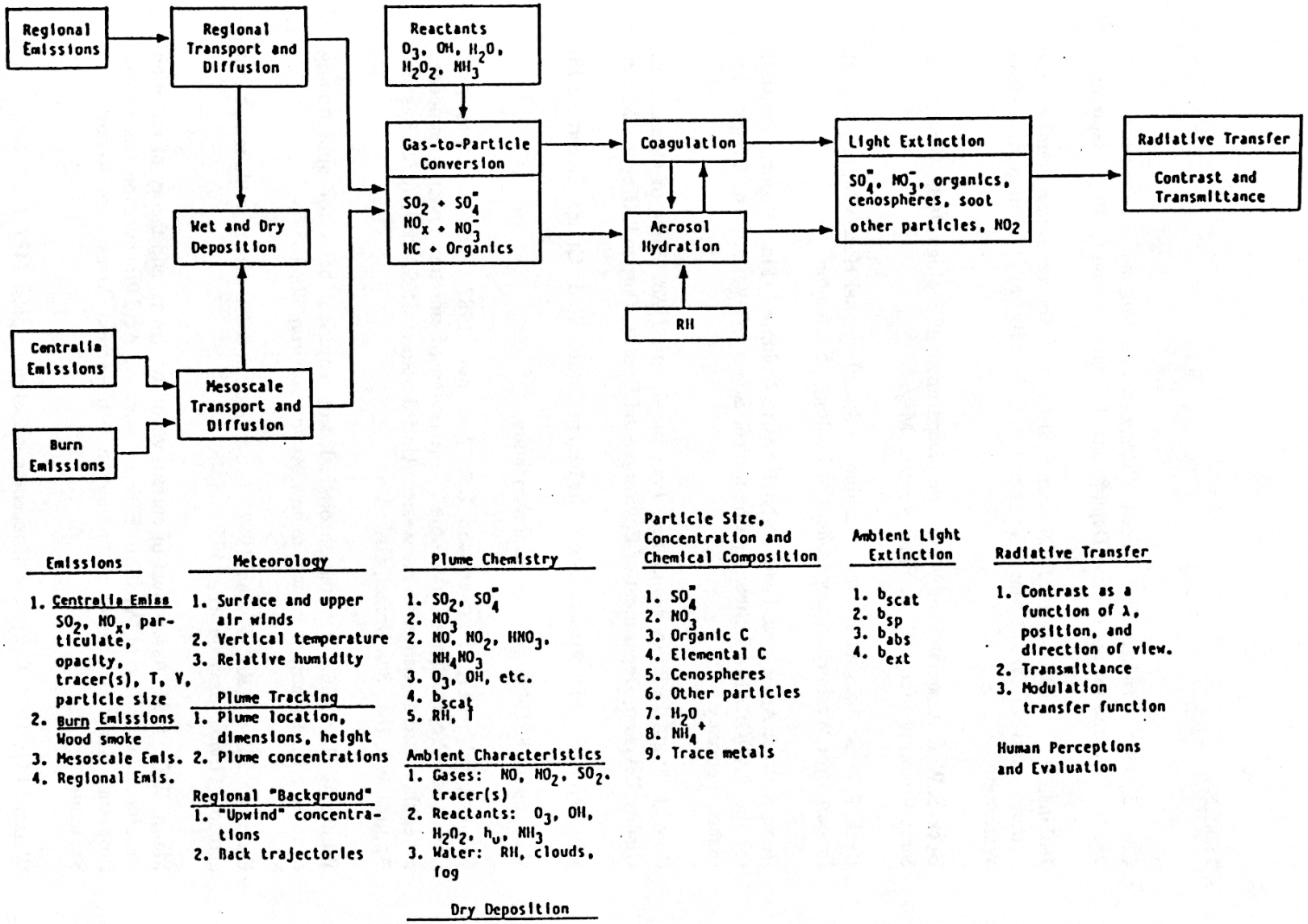

Figure 1-4. Categories of measurements and cause-effect relationships.

REFERENCES

1. Core, J., 1985. *Visibility in Oregon's Wilderness and National Park Lands (1982-1984)*. Prepared for the Oregon Department of Environmental Quality. September.

2. Jenkins, P.G., 1982. *Aerosol Characterization and Source Determination in a Federal Class I Area*. Master's Thesis. University of Washington, Seattle, Washington.

3. Beck, R.W. & Associates and Washington Department of Ecology, 1983. Washington State Visibility Study. 1982 Final Report. May.

4. Beck, R.W. & Associates, 1984. Washington State Regional Haze/Visibility Report. Prepared for Washington Department of Ecology. September.

5. Beck, R.W. & Associates, 1986. PANORAMAS Volume 1, Final Report. Prepared for the PANORAMAS Steering Committee, States of Washington, Oregon, and Idaho. February.

6. Core, J., 1989. *Visibility Monitoring Program Report 1988*. State of Oregon, Air Quality Division, Department of Environmental Quality, Portland, Oregon. March.

7. Van Haren, F., 1989. Presentation to the Mount Rainier Study Group. December 13.

8. Van Haren, F. 1990. Personal communication.

9. Chinkin, L.R., D.A. Latimer, and L.A. Mahoney, 1987. Western States Acid Deposition Project Phase I: Volume 2 -- A review of emission inventories needed to regulate acid deposition in the western United States. SYSAPP-87/072. Systems Applications, Inc., San Rafael, CA. May.

10. White, W.H., 1986. On the theoretical and empirical basis for apportioning extinction by aerosols: a critical review. *Atmos. Environ.* **20**, 1659.

11. Ouimette, J.R. and R.C. Flagan, 1982. The extinction coefficient of multicomponent aerosols. *Atmos. Environ.* **16**, 2405.

12. Malm, W.C., 1991. Assessment of current visibility effects, and theory of radiation transfer and visibility. In "Visibility Effects." National Acid Precipitation Assessment Program's State of Science/Technology and Integrated Assessment Report #24. September.

13. Watson, J.G., 1984. Overview of receptor model principles. *JAPCA*, **34**, 620.

14. Gordon, G.E., 1980. Receptor models. *Environ. Sci. Technol.* **14**, 795.

15. Cooper, J.A., J.G. Watson, 1980. Receptor-oriented methods of air particulate source apportionment. *JAPCA*, **30**, 1116.

16. Currie, L.A., R.W. Gerlach, C.W. Lewis, et al., 1984. Interlaboratory comparison of source apportionment procedures: results for simulated data sets. *Atmos. Environ.*, **18**, 1517.

17. Dzubay, T.G., R.K. Stevens, W.D. Balfour, et al., 1984. Interlaboratory comparison of receptor model results for Houston aerosol. *Atmos. Environ.*, **18**, 1555.

18. Watson, J.G. and N.F. Robinson, 1984. A method to determine accuracy and precision required of receptor model measurements. *APCA Quality Assurance in Air Pollution Measurements.* Pittsburgh, PA.

19. Malm, W.C., H. Iyer, J.Watson, et al., 1989. Survey of a variety of receptor modeling techniques. In *Proceedings of the AWMA/EPA International Specialty Conference on Visibility and Fine Particles.* Estes Park, CO.

20. Malm, W.C. and H.K. Iyer, 1989. Application of tracer mass balance regression to WHITEX data. In *Proceedings of the AWMA/EPA International Specialty Conference on Visibility and Fine Particles.* Estes Park, CO.

21. Hopke, P.K., 1985. Receptor modeling in environmental chemistry. In *Chemical Analysis*, John Wiley & Sons, New York, NY, 76.

22. Whitmore, J.B., W.C. Malm, and H.K. Iyer, 1991. Sensitivity analysis of tracer mass balance regression. In *Proceedings of the 84th Annual Meeting of the Air & Waste Management Association*, Vancouver, B.C.

23. Dzubay, T., R.K. Stevens, G.E. Gordon, et al., 1988. A composite receptor method applied to Philadelphia aerosol. *Environ. Sci. & Technol.*, **22**, 1.

24. Lewis, C.W. and R.K. Stevens, 1985. Hybrid receptor model for secondary sulfate from an SO_2 point source. *Atmos. Environ.*, **19**, 917.

25. Stevens, R.K. and C.W. Lewis, 1987. Hybrid receptor modeling. In *Extended Abstracts for the Fifth Joint Conference on Applications of Air Pollution Meteorology*, APCA, American Meteorological Society, Boston, MA.

26. Watson, J., editor, 1989. *Transactions, Receptor Models in Air Resources Management.* Air & Waste Management Association, Pittsburgh, PA.

27. Malm, W. C., K. Gebhart, D. Latimer, T. Cahill, R. Eldred, R. Pielke, R. Stocker, and J. Watson, 1989. National Park Service Report on the Winter Haze Intensive Tracer

Experiment. Draft Final Report. National Park Service. Fort Collins, CO. December.