OVERVIEW AND SYNOPSIS

The Pacific Northwest Regional Visibility Experiment Using Natural Tracers (PREVENT) was carried out during the summer of 1990. The objectives of the study were to:

- determine the spatial and temporal patterns of visibility reducing aerosol concentrations;

- determine estimates of light extinction budgets for the summer period for Mount Rainier and North Cascades National Parks;

- apportion summertime haze constituents to general source types found in the Northwestern United States and Southwestern Canada; and

- develop estimates of the amount of haze that is natural and man-made.

The report consists of eleven chapters. Chapter 1 describes the study area in terms of its geographic extent and known emissions. It also includes a general discussion of the data analysis techniques and protocols. In Chapter 2 the field monitoring program is presented. It includes a discussion of the various types of instrumentation used as well as the geographic locations of each of the monitors. Chapters 3 and 4 contain a discussion on quality assurance protocols and measurement uncertainties for the optical and aerosol monitors.

The aerosol sampling network consisted of three primary and 31 secondary sites. The three primary sites were the Mount Rainier sites at Tahoma Woods and Paradise and the North Cascades site at Marblemount. At the primary sites the major aerosol species contributing to visibility reduction were monitored on a 12-hour sampling interval starting at 0800. The aerosol types included sulfur and sulfate ions, nitrate ions, organic and light-absorbing carbon, optical absorption, fine (0.0-2.5 μm) and coarse (0-10.0 μm) gravimetric mass, and soil related elements. Additionally, the filters were analyzed using high sensitivity techniques for trace elements. At the 31 secondary sites samples were collected in the 0.0-2.5 μm size range on Teflon substrates. The samples were analyzed for gravimetric mass, elements, and optical absorption. From these measurements it was possible to derive estimates of sulfates, organic mass, atmospheric absorption, and fine soil concentrations.

At two sites, Marblemount and Tahoma Woods, integrating nephelometers were modified to make near ambient measurements of the atmospheric scattering coefficient while at the third site, Paradise, an unmodified instrument was operated. Unmodified nephelometer measurements were made at five additional sites. Five automatic 35 mm and 8 mm time lapse camera systems were installed specially for this study while another ten 35 mm camera sites were operated as part of other ongoing monitoring programs.

The full suite of measurements made at the primary sites allowed for a number of analyses to be carried out. The aerosol data were used to develop estimates of sulfate acidity. These data when combined with nephelometer measurements allowed for an estimation of the relative contribution of each aerosol species to extinction. Finally, the high sensitivity trace element data were used to develop estimates of how various source types contributed to aerosol loads.

The spatial distribution of various aerosol species derived from the 31 secondary sites helped in developing an understanding of the location and source strength of sources contributing to elevated particle concentrations.

Chapter 5 is a weather summary for the Pacific Northwest for the time period of July 6 to September 6, 1990. The weather vacillated between two characteristic patterns. The first of these patterns was associated with high surface pressures, light northerly winds, and clear skies. The second was associated with lower pressure, southerly winds, and overcast.

Operating nephelometers without regard to the inadvertent heating of the aerosol sample as it passes through the sampling chamber can result in significant underestimates of the ambient scattering coefficient. In Chapter 6, the effects of inadvertent sample heating are explored. Also, presented in Chapter 6 is a "climatology" of aerosols and their related effect on extinction.

It is estimated that operating the commercially available nephelometers on the western side of the North Cascades Mountain Range without regard to inadvertent heating of the aerosol will cause the ambient scattering coefficient to be underestimated by about a factor of seven. Even after substantial effort to modify the nephelometers to make ambient scattering measurements, it is estimated that the scattering coefficient was still underestimated by about a factor of two.

At both Tahoma Woods and Marblemount the average fine mass concentration is about 8-10 $\mu g/m^3$ with maximums of 20-30 $\mu g/m^3$. About 20-30% of the measured gravimetric fine mass is estimated to be associated with water. Of the measured PM_{10} (< 10.0 μm) about half are fine particles.

Figure 1 is a summary, presented in the form of a pie chart, of the fraction of the fine mass associated with the various aerosol species. Organics made up the largest fraction of fine mass at about 50% while light absorbing carbon contributed another 10%. Together, carbon- related aerosols were responsible for nearly 60% of the measured fine mass. Sulfates, presumed to be in the form of ammonium sulfate, contributed about 30% of the fine mass. Surprisingly, nitrates contributed only about 5% of the mass with soil making up about another 7%.

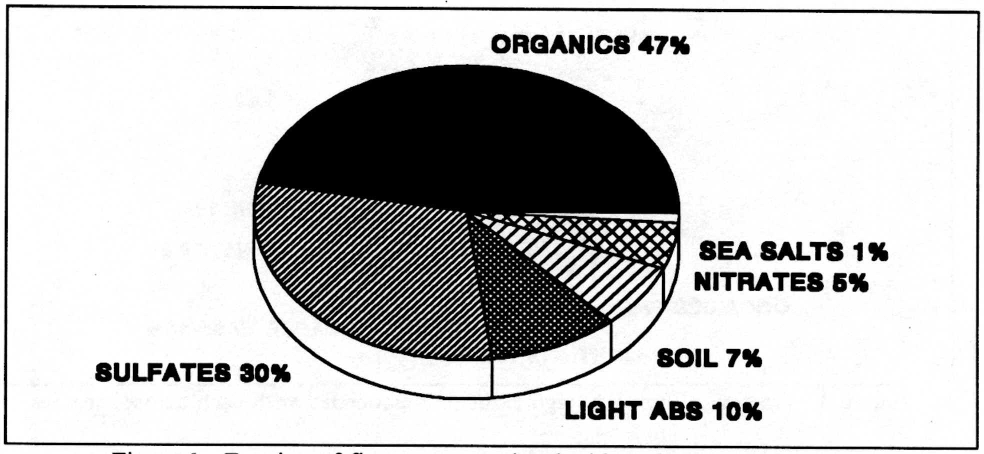

Figure 1. Fraction of fine mass associated with various aerosol species.

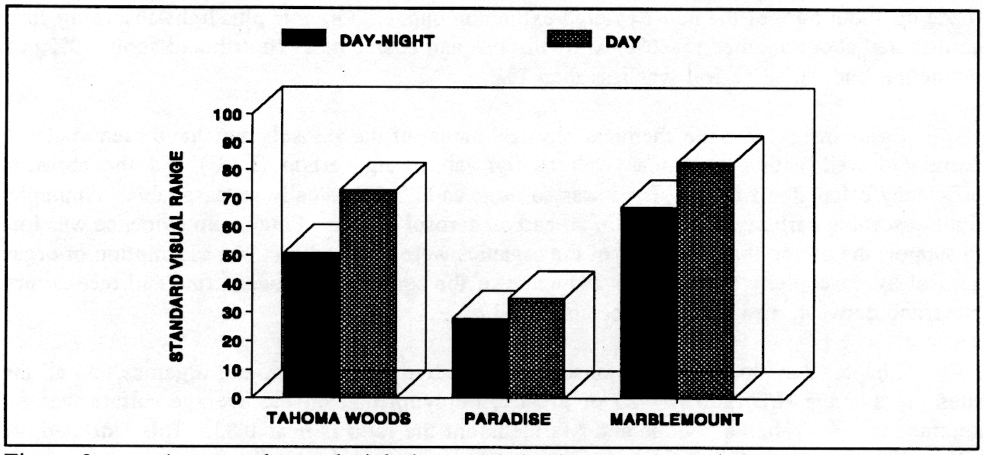

Figure 2. Average day and nighttime, and daytime standard visual range estimates for Tahoma Woods, Paradise, and Marblemount.

The average day and nighttime standard visual range for the primary monitoring sites is presented in Figure 2. The dark bars are averages of day and nighttime standard visual range while the lighter bars are daytime only visual ranges. Daytime standard visual ranges are calculated using the 12-hr samples starting at 0800. Daytime visual ranges are higher primarily because the relative humidity is lower during daylight hours. The best visibility is found at the North Cascades monitoring site. At Tahoma Woods, the low elevation Mount Rainier site, visual range is somewhat less while at the high elevation Mount Rainier site, Paradise, average visual range is estimated to be the lowest. However, the Paradise data set is abbreviated (shorter time span) and therefore is not representative of the same time period as the other two sites.

On the average, blue sky scatter or Rayleigh scattering, makes up about 10-20% of the overall visibility or extinction budget. Figure 3 is a graphical summary of the average contribution of aerosol species to non-Rayleigh extinction. Non-Rayleigh extinction is all extinction other than blue sky scatter.

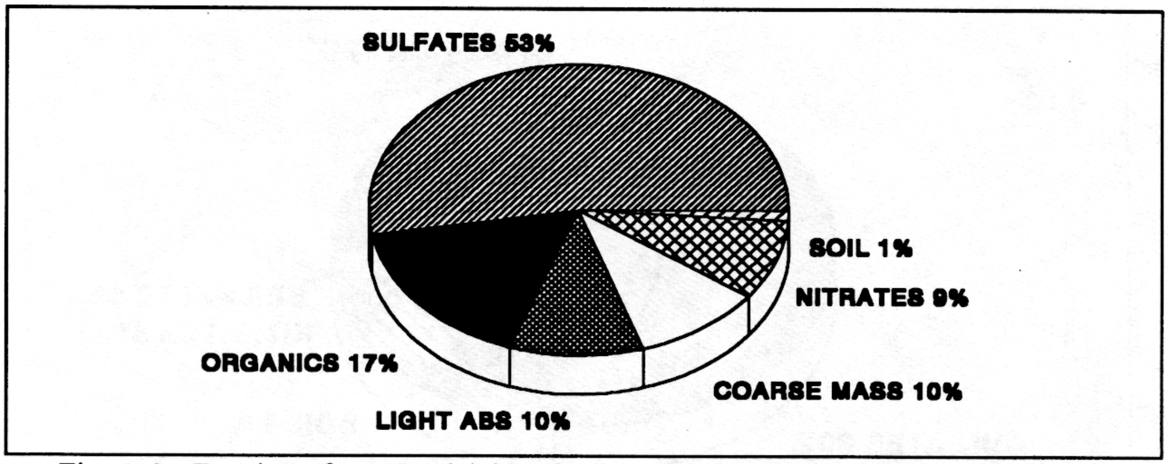

Figure 3. Fraction of non-Rayleigh extinction associated with each aerosol species.

Even though sulfate accounted for only 20-30% of the fine mass it was estimated that it made up about 50% of the non-Rayleigh extinction budget. Organic plus light-absorbing carbon contributed about another 15-20%, while nitrates and coarse mass contributed about 10% to the extinction budget. Fine soil was less than 1%.

Some insight into the chemical-physical nature of the aerosols may have been made. b_{abs} correlated well with organics as well as light-absorbing carbon (LAC) and the absorption efficiency calculated from b_{abs}/LAC was so large as to be physically unreasonable. Apparently, light-absorbing carbon was present in all carbon aerosol species. Finally, no evidence was found to support the notion that a fraction of the organics were hygroscopic. An assumption of organic aerosol hygroscopicity resulted in a reduction of the correlation between fine and reconstructed mass and between measured and reconstructed b_{scat}.

Chapter 7 explores the relationship between hydrogen, sulfur, and organics. At all three sites the average H/S ratio is 0.25 or greater, implying that on the average sulfate was fully neutralized. At Tahoma Woods and Marblemount the ratio is near 0.33. This "inflated" H/S ratio is suggestive of an organic-sulfur multicomponent aerosol. At all three sites there are a few time periods where the H/S ratio suggests acidic aerosols.

Chapters 8, 9, 10, and 11 taken together help understand the sources of visibility reducing aerosols. In Chapter 8, the relationships between visibility reducing aerosols and various trace elements that were grouped using factor analysis were explored. This approach was strictly empirical. In Chapter 9, chemical mass balance (CMB) was used to apportion those elements that were found to be related to visibility reducing aerosols to source types. This analysis gives some insight into sources contributing to visibility reduction. Finally, in Chapter 9, the relative source strengths were used as independent variables in an ordinary least square regression while visibility reducing particles were used as dependent variables. This method is referred to as CMB regression analysis. In this way the average contribution of source types was linked to visibility reduction.

Together the empirical and CMB regression analyses yielded insight into which sources contributed to elevated levels of visibility reducing aerosols. In Chapter 10, back trajectory analysis was used to develop the frequency with which specific sources within general source categories contributed to elevated particulate levels. In Chapter 11, the spatial distribution of particle concentrations yielded further insight into which sources contributed to particle concentrations. For instance, it was expected that the highest concentrations of organics would be found near a forest fire with decreasing concentrations as one moves radially away from the particle source. Each of the major visibility reducing aerosol types were investigated using these various techniques.

The source attribution analyses focused on the Tahoma Woods and Paradise monitoring sites because it was only at these sites that high sensitivity trace element analysis was available.

Sulfur

Sulfur was the single largest contributor to visibility reduction. From a statistical point of view it was linked to silicon, bromine, and selenium at Tahoma Woods and selenium and high temperature elemental carbon at Paradise. At Tahoma Woods, bromine explained the largest variance in the sulfur data set while at Paradise selenium explained the largest variance. Bromine was associated with about a third of the sulfur at Tahoma Woods while at Paradise selenium was associated with about a third of the sulfur. The CMB analysis apportioned between 75-79% of bromine to transportation and 78-90% of the selenium to coal-fired power plants. At Tahoma, Woods the CMB regression analysis apportioned about another 20% of the sulfur to transportation activity. CMB regressions did not yield statistically significant regression coefficients at Paradise and in general did not yield model fits that were comparable to those at Tahoma Woods.

Back trajectories were used to determine how often the coal-fired power plant was associated with the Centralia Power Plant. Figure 4 shows a temporal plot of sulfur, the period of time that back trajectories passed over the Centralia Power Plant, and the strength of the CMB predicted coal-fired power plant signature for Paradise and Tahoma Woods. Specific time periods of interest are identified by numbers 1-11.

First, notice that sulfur levels at Paradise were episodic. There were periods where concentrations were high followed by time periods where sulfur levels were near zero. On the other hand, sulfur levels at Tahoma Woods were less episodic and tended to stay elevated much of the time. Most sulfur episodes at Paradise were associated with air masses that passed over the Centralia Power Plant. These time periods are shown by shaded areas in Figure 4. The CMB analysis also predicted a coal-fired power plant "hit" four of the six times that back trajectory analysis showed the Centralia Power Plant impacting Paradise. There were, however, time periods when sulfur at Paradise was not associated with the Centralia Power Plant. The episodes starting July 27 (episode number 2) and August 28 (episode number 10) were apparently associated with coal-fired power plant emissions (high relative CMB strength), but not with air masses coming from Centralia. Also, the largest sulfur episode starting on August 12 (episode number 7) was apparently associated with two distinctly different sources. The first half of the episode (August 12-14) showed a coal-fired power plant signature and back trajectories from Centralia.

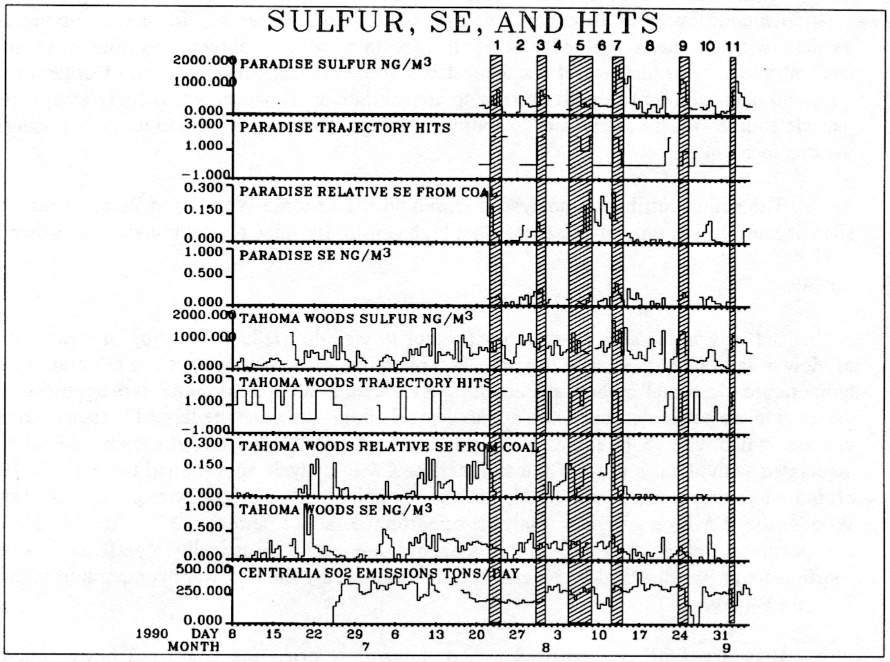

Figure 4. A temporal plot of sulfur, frequency of trajectory intersections with the Centralia, relative strength of coal-fired power plant signature, and Se at Paradise and Tahoma Woods.

During the second half of the episode CMB analysis showed a near zero impact from coal-fired power plants and back trajectories from the southwest well away from the Centralia Power Plant.

At Tahoma Woods, the relationship between back trajectories that pass over Centralia and elevated sulfur was not as close as at Paradise. Sulfur was more uniformly elevated at Tahoma Woods and did not show the same strong episodic nature it did at Paradise. However, the time period corresponding to the largest sulfur episode at Paradise (August 12-14 episode number 7) was also the largest sulfur episode at Tahoma Woods. As at Paradise, the CMB analysis predicted the largest relative coal-fired power plant impact and the back trajectories passed over the Centralia Power Plant. The second half of the episode (August 15-16) did not appear to be coal-fired power plant related.

The spatial trend analysis presented in Chapter 11 tends to confirm the back trajectory analysis. Spatial trends were explored using empirical orthogonal function analysis (EOF). An EOF analysis simply groups those spatial patterns that tend to occur over and over again into separate and distinct spatial patterns. For sulfur there are three distinct patterns (See Figures 5, 6, and 7). The first EOF, Figure 5, corresponds to high sulfur concentrations around the Seattle-Tacoma urban area, which is likely associated with urban-transportation sources of sulfur. The second EOF, Figure 6, is associated with west-east transport with a build up of sulfur along the western slope of the Cascades as the flow becomes blocked by the mountain range. The third EOF, Figure 7, corresponds to transport from Canada into the study area.

Figure 8 shows the strength of each pattern as a function of time. Comparing the time line of sulfur at Paradise and EOF 2 (see Figure 8) shows that EOF 2, west-east transport, is associated with almost every sulfur episode at Paradise. This is consistent with back trajectories passing over the Centralia Power Plant (west-east transport). On the other hand, sulfur levels at Tahoma Woods are combinations of EOF 1 and EOF 2, especially for the time period from July 20 - August 11. The episode starting on August 12 that occurs at both Tahoma Woods and Paradise is associated with EOF 2. Again this is consistent with the CMB and back trajectory analyses which suggests that the first half of the episode is linked to Centralia emissions.

The EOF analysis does give some insight into the origin of sulfur at Marblemount. EOF 3 is associated with sulfur transport from Canada into northern Washington. Because the temporal plot of sulfur at Marblemount is very similar to the temporal variation of EOF 3 it can be inferred that much of the sulfur at Marblemount is from transboundary transport of sulfur from Canada.

In summary, the source apportionment analysis presented in Chapters 8 and 9 showed that sulfur was statistically linked to coal-fired power plants and urban emissions. The trajectory analysis combined with spatial trends in sulfur concentrations (Chapters 10 and 11) tended to confirm an urban-transportation (Seattle-Tacoma area) and a coal-fired power plant (Centralia Power Plant) influence on sulfur concentrations at Mount Rainier. Furthermore, at Paradise the correlation between back trajectories that pass over Centralia and elevated sulfur suggested that the Centralia Power Plant may be a major contributor to elevated sulfur levels at Paradise. There were also time periods where it was clear that coal-fired power plants other than Centralia contributed to sulfur at both monitoring sites. However, the statistical relationships were not judged to be "strong" enough to attempt quantitative apportionment of sulfur to specific sources.

Nitrates

Nitrates contributed about 10% of the visibility reduction at North Cascades and Mount Rainier National Parks. At both Paradise and Tahoma Woods, nitrates were most closely associated with the element sodium, and CMB analysis suggested that most of the sodium found at these sites was linked to either pulp and paper mill activity or the lime-kiln profile. The CMB regression analysis yielded a model which was strong (r^2=0.79) at Tahoma Woods but weak at Paradise. It was estimated that between 20-50% of the nitrates were related to pulp and paper mill or lime-kiln activity; 20-30% with burning, while coal-fired power plants and transportation contributing to nitrates were in the 10-20% range.

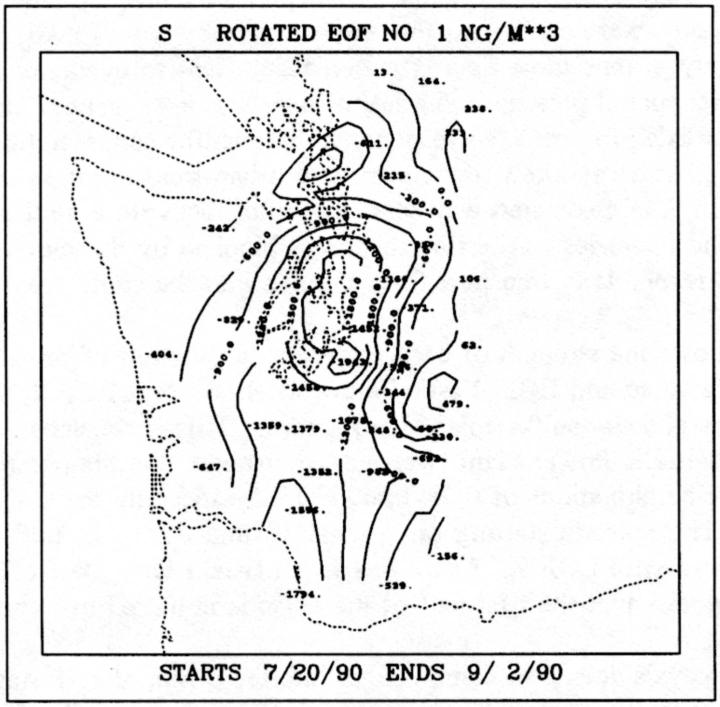

Figure 5. Spatial pattern of sulfur for EOF1.

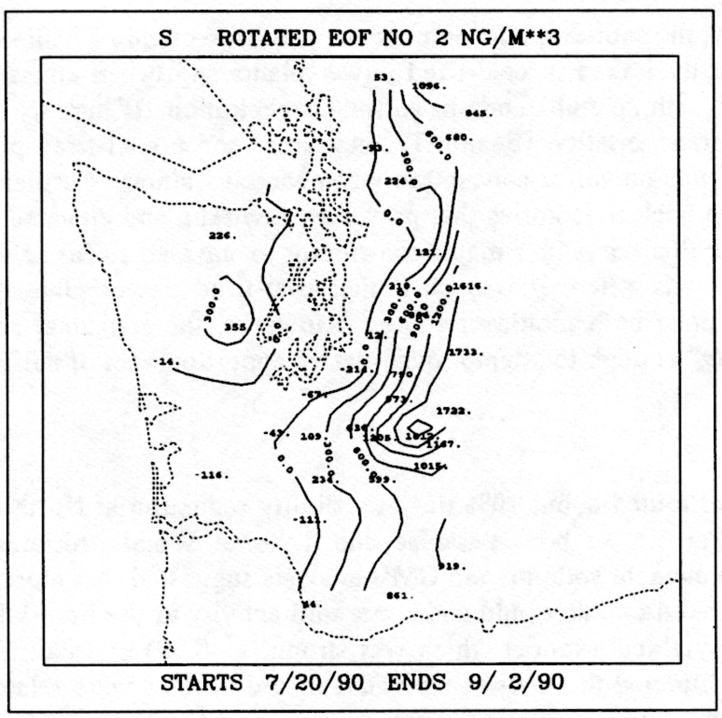

Figure 6. Spatial pattern for sulfur for EOF2.

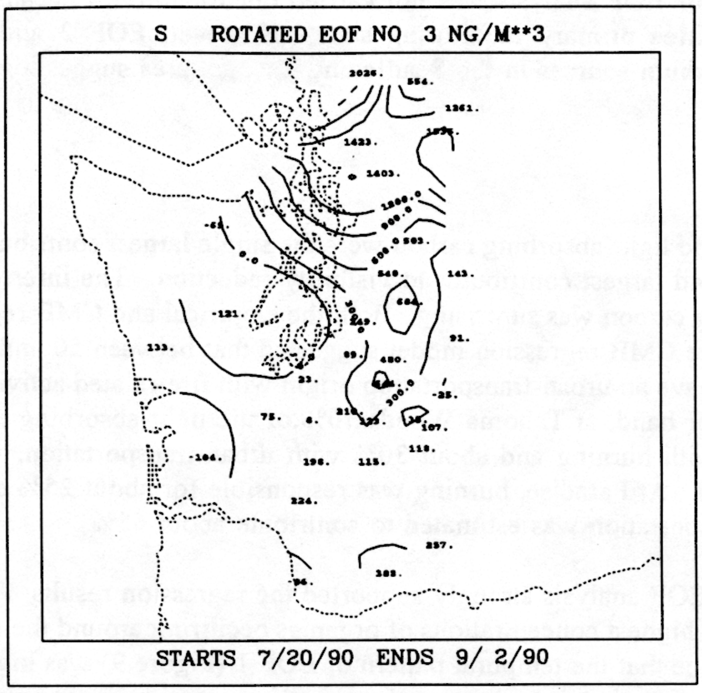

Figure 7. Spatial pattern for sulfur for EOF3.

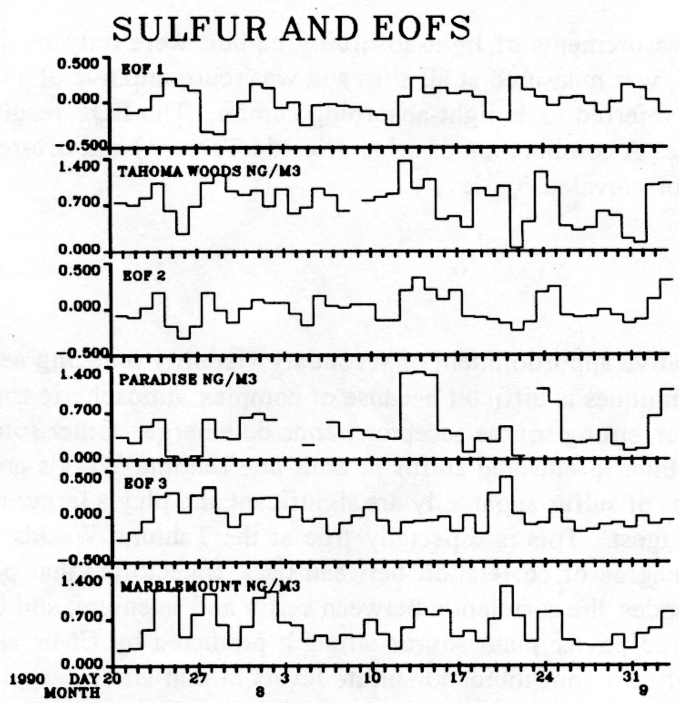

Figure 8. Temporal plot of sulfur at Tahoma Woods, Paradise, and Marblemount and the relative strength of EOF1, EOF2, and EOF3.

Trajectory or EOF analysis was not carried out for nitrates because nitrates were only measured at the three primary monitoring sites. However, EOF 2 and EOF 3 for sodium suggested large sodium sources in the Seattle and Tacoma area suggesting pulp and paper mill emissions.

Organics

Organics and light-absorbing carbon were the single largest contributors to measured fine mass and the second largest contributor to visibility reduction. The inferred origin of organics and light absorbing carbon was surprising. Both the empirical and CMB regression models have high r^2 values. The CMB regression model suggested that between 50 and 60% of the organics at Mount Rainier have an urban-transportation origin with fire-related activity accounting for 20-40%. On the other hand, at Tahoma Woods 70% of the light-absorbing carbon was estimated to be associated with burning and about 30% with urban-transportation, while at Paradise the trend was reversed. At Paradise, burning was responsible for about 25% of the light-absorbing carbon while transportation was estimated to contribute about 65%.

Again the EOF analysis strongly supported the regression results. The spatial pattern of EOF 1 showed the highest concentrations of organics occurring around the urban areas of Seattle and Tacoma. Notice that the temporal pattern of EOF 1 (Figure 9) was highest during the week with minimums on weekends. Apparently, more organics were generated along the urban corridor during the working week. EOF 3 and EOF 4 seem to be associated with fire-related activity in that they were isolated in space and peak only on a few days (see Figures 10, 11, and 12).

Separate measurements of light-absorbing carbon were only made at the three primary sites. However, b_{abs} was measured at all sites and was representative of a combination of organic and what has been referred to as light-absorbing carbon. The EOF results for b_{abs} were similar to those for organics. A combination of urban (Seattle-Tacoma) and fire-related activity appeared to be responsible for elevated b_{abs} levels.

SUMMARY

The quantitative apportionment of secondary visibility reducing aerosols to sources using source-receptor techniques is difficult because of complex atmospheric transport, dispersion, and chemistry. However, strong source-receptor trends do emerge. Other sources than the Centralia Power Plant contribute to elevated sulfur at both the Tahoma Woods and Paradise monitoring sites. Urban sources of sulfur apparently are significant and play a larger role than their emission strengths would suggest. This is especially true at the Tahoma Woods location. However, at Paradise the high degree of correlation between back trajectories that pass over Centralia and elevated sulfur episodes, the correlation between sulfur and selenium, and the association between the relative coal-fired power plant source strength predicted by CMB analysis all suggest that Centralia is a significant contributor to sulfate levels at that site.

The largest source of nitrates appeared to be pulp and paper mill activities with contribution from coal-fired power plants, transportation, and burning.

Most of the organic carbon was associated with transportation-related emissions in the Seattle-Tacoma urban areas while more of the light-absorbing carbon was associated with fire-related activity.

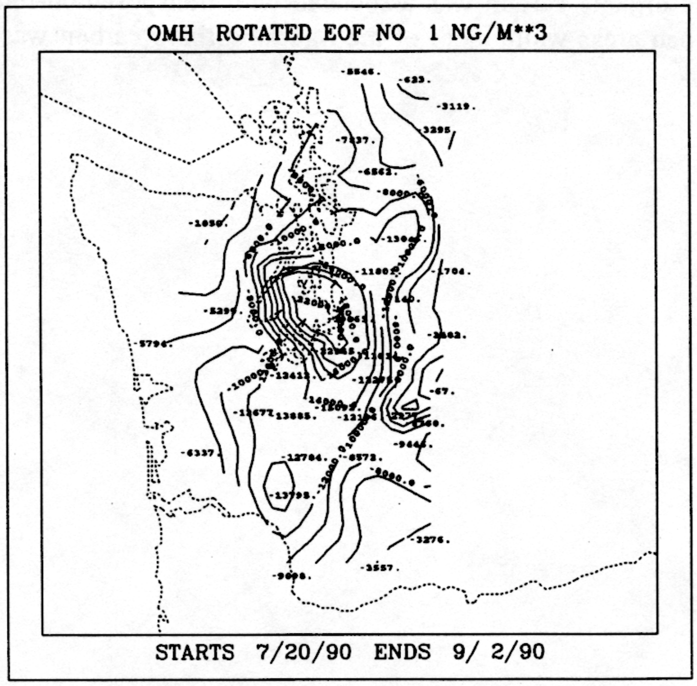

Figure 9. Rotated EOF 1 for OMH explaining 38.7% of the variance.

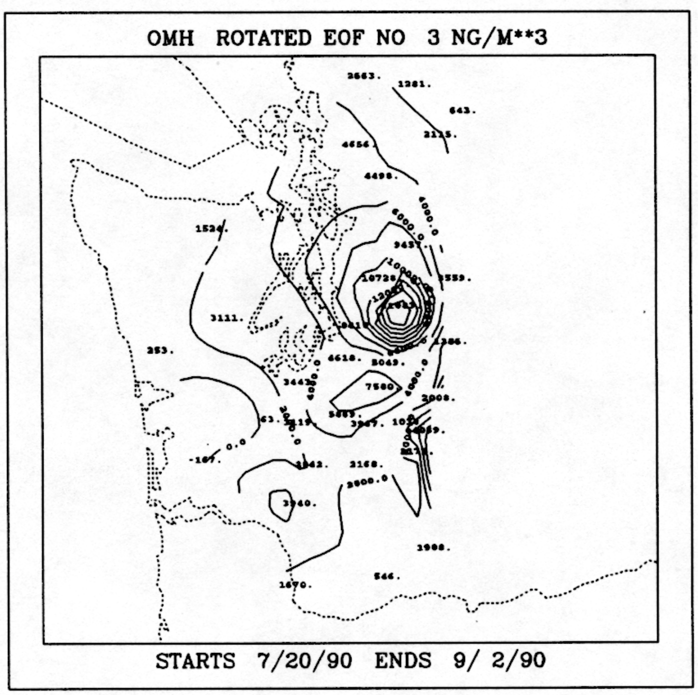

Figure 10. Rotated OMH EOF 3 explaining 11.8% of the variance. Contour interval is 2000 ng/m^3.

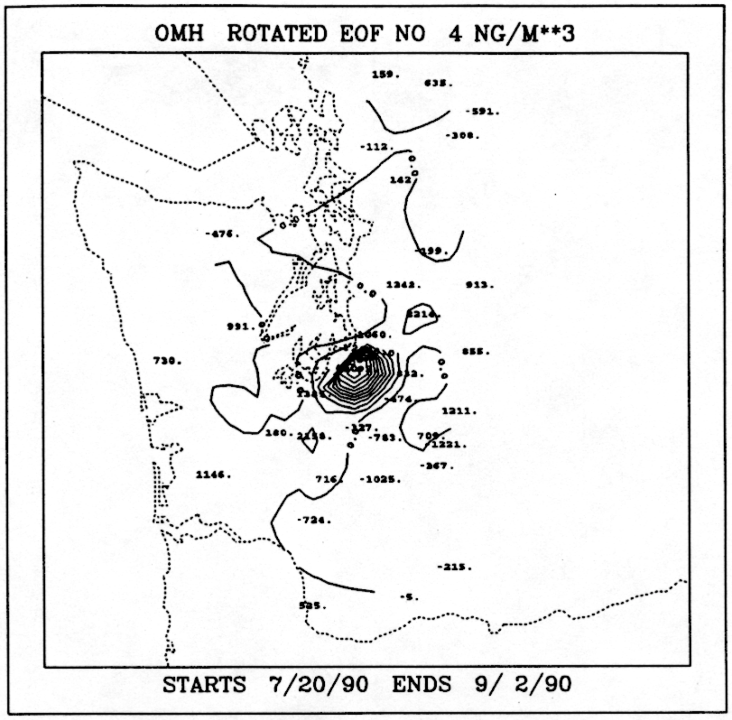

Figure 11. Rotated OMH EOF 4 explaining 9.2% of the variance. Contour interval is 2000 ng/m^3.

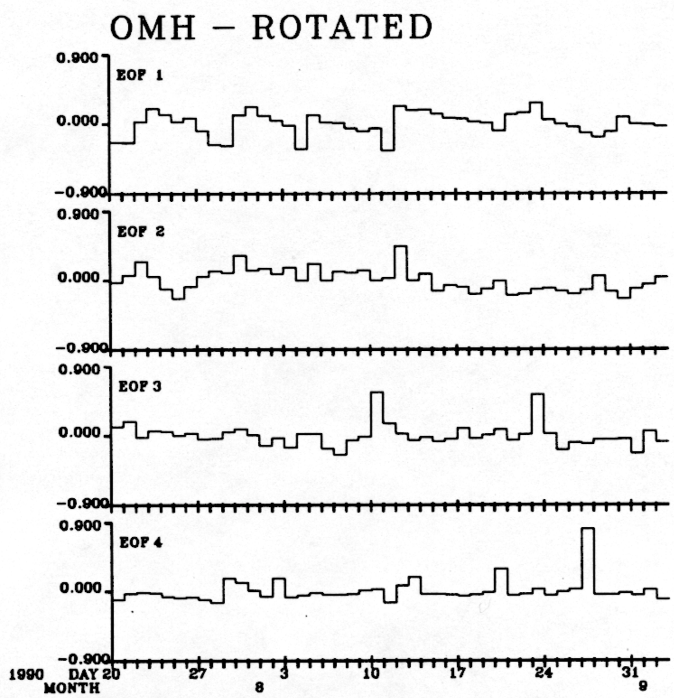

Figure 12. Time factors for first four rotated EOFs for OMH.