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CORRELATIONS AMONG VISUAL RANGE,
AEROSOL SULFATE LEVEL, AND RELATIVE HUMIDITY
IN THE MOUNTAINS OF NORTHWESTERN NORTH CAROLINA

A Thesis
by
DAVID CHARLES WILSON

Submitted to the Graduate School
Appalachian State University
in partial fulfillment of the requirements for the degree of
MASTER OF ARTS

August 1982

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ABSTRACT

CORRELATIONS AMONG VISUAL RANGE, AEROSOL SULFATE
LEVEL, AND RELATIVE HUMIDITY IN THE MOUNTAINS
OF NORTHWESTERN NORTH CAROLINA. (August 1982)

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A 7-month, USEPA-sponsored field study was conducted in the Appalachian mountains of northwestern North Carolina to investigate the effects of ambient sulfate aerosol concentration on reduced visibility in that region. Aerosol was collected on filters with a dichotomous sampler, visual range was determined photographically from a known set of mountain peaks, and relative humidity was monitored continuously with a human hair hygromograph. A high correlation was found between fine particulate sulfate mass concentration and visual range. The correlation between total fine particle mass and visual range was reasonably high, with a corrected slope that was comparable to a semi-empirical value calculated by Dzubay. Mean sulfate mass (as $(\text{NH}_4)_2\text{SO}_4$) averaged 38% of the total fine particle mass and decreased from summer to winter.

Acknowledgements

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Special thanks go to my cohort, Steve Breiner, for useful thesis discussions, and to Sherry Fonvielle for preparation of the manuscript.

My warmest appreciation is given to the members of my family for all the years of their love and moral support.

Dedication

These pages are dedicated with love to my fiancée, Lynn,
with hopes for a wonderful lifetime together.

TABLE OF CONTENTS

	<u>Page</u>
List of Tables	ix
List of Figures	x
I. INTRODUCTION AND BACKGROUND	1
A. Introduction	1
B. The Nature of Sulfate Aerosol	2
C. Fine Particle Optics	4
D. Measurement of Visual Range	5
II. EXPERIMENTAL PROCEDURE	7
A. Visual Range Measurements	7
B. Aerosol Measurements	7
C. Relative Humidity Measurements	12
III. RESULTS AND DISCUSSION	13
IV. CONCLUSION	25
REFERENCES	27
VITA	29

LIST OF TABLES

<u>Table</u>	<u>Page</u>
I. Master Data Table	14,15
II. Supplemental Data Table	16
III. Summary of Recent Aerosol Studies	19

LIST OF FIGURES

<u>Figure</u>	<u>Page</u>
1. Schematic view of the Beckman dichotomous sampler (14)	9
2. Cross-sectional view of virtual impactor designed by Loo et al., 1978 (14)	10
3. Dionex ion chromatograph flow scheme illustrating anion analysis (15)	11
4. Fine particulate sulfate concentration vs. inverse visual range, with relative humidity indicated beside each data point	17
5. Total fine particle mass concentration vs. inverse visual range, with relative humidity indicated beside each data point	21
6. Weekly average bar graph for fine particulate sulfate and total fine particle mass concentrations through the study period	24

CHAPTER I
INTRODUCTION AND BACKGROUND

A. Introduction

One value of air clarity is the aesthetically pleasing property of allowing one to see for long distances with an unimpeded view. Visibility of significant vistas is considered an important part of the natural heritage for national parks and wilderness areas. In recognition of visibility as a natural resource worthy of being preserved, Congress in 1977 enacted the Clean Air Act Amendments establishing national visibility goals. This has spurred numerous investigations into the sources of air pollutants and the effects of impaired visibility which result from these pollutants (1).

The largest class of air pollutants has as its primary characteristic the presence of particles in the atmosphere. According to Lodge et al. (2), the most important non-health effect of airborne particulate matter at its usual concentration levels is its impact on visibility. Recent investigations (3-9) have indicated that of the major constituents of the total suspended mass, sulfate (SO_4^{2-}) with its associated cations contributes most to light extinction. Also, aerosol formation by the

growth of these particles as relative humidity increases has been shown to result in a further decrease in visibility.

The present study was designed to investigate sulfate aerosol in the Appalachian mountains of northwestern North Carolina. The techniques used were patterned after similar aerosol studies conducted in two other areas of the southern Appalachians: (a) the Great Smoky Mountains study by Stevens et al. (4), and (b) the Shenandoah Valley studies by Ferman et al. (3) and by Stevens et al. (5). The study was conducted in cooperation with the Environmental Protection Agency at Research Triangle Park, N. C. (EPA-RTP) and took place from mid-July, 1981 through February, 1982 in and around Boone, N. C. Boone is a small (population 20,000) university town on the eastern slopes of the southern Appalachians, roughly equidistant from the Great Smoky Mountains National Park to the southwest and the Shenandoah National Park to the northeast. By monitoring ambient particulate sulfate concentrations, visual range, and relative humidity in the study area, an attempt was made to correlate sulfate level with visibility, while noting the relative humidity effect. Attention was also given to the contribution of sulfate to the total mass of fine particles (<3.5 μm in diameter), the correlation between total fine particle mass and visibility, and to sulfate level trends throughout the seven month study period.

B. The Nature of Sulfate Aerosol

Sulfate occurs in the atmosphere as the final oxidation product of sulfur dioxide (SO_2) gas originating mainly from

anthropogenic sources, primarily from the burning of coal and petroleum products and ore smelting. In the United States, roughly two-thirds of the airborne SO_2 is produced by the electric utilities and one-fourth by industry (10). Homogeneous photooxidation of SO_2 by oxidizing radicals such as hydroxyl constitutes a major route for sulfate formation, with the primary form initially produced as sulfuric acid (H_2SO_4). Heterogeneous conversion routes include the oxidation of SO_2 in the liquid phase by catalysis or strong oxidant, and the surface-catalyzed oxidation of SO_2 upon collision with solid particles (11). Sulfuric acid aerosol is in turn neutralized wholly or in part by atmospheric ammonia to form ammonium bisulfate (NH_4HSO_4) or ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$). Hence, the sulfate aerosol may be characterized as a mixture of H_2SO_4 , NH_4HSO_4 , and $(\text{NH}_4)_2\text{SO}_4$, with relative abundances depending upon atmospheric conditions. Since these particles are hygroscopic, this mixture contains associated water molecules in a range of concentrations dictated by the relative humidity (2).

Suspended particles in the atmosphere range in size from about 0.001 to 100 μm in diameter, and exist in a bimodal distribution (2). The particle size for sulfate aerosol falls predominantly in the mode below about 3.5 μm diameter within the population called the fine particulate fraction. Of the constituents of the fine particulate fraction, sulfate with its associated cations has been shown to be the major species present, comprising as much as 61% of the particle mass (4). Particles in this size range are not readily removed by settling or meteorological processes. Coagulation

occurs only slowly so that the aerosol tends to be very persistent in the atmosphere, having a typical airborne lifetime of many days (2, 10). Therefore, particulate sulfate may be carried hundreds or thousands of miles by prevailing winds before it is brought down as acid precipitation or deposited as dry particles (10). As a result, ambient sulfate aerosol does not merely originate from local sources.

C. Fine Particle Optics

According to the Beer-Lambert law, transmission of light through the atmosphere may be described as (12)

$$\frac{\Delta I}{I} = -b_{\text{ext}} \Delta x \quad (1)$$

where the extinction coefficient, b_{ext} , of the atmosphere determines the change in intensity, ΔI , of light traversing a path of length Δx . This extinction coefficient can be expressed as a sum of scattering and absorption components for both gases and particles (6)

$$b_{\text{ext}} = b_{\text{sg}} + b_{\text{sp}} + b_{\text{ag}} + b_{\text{ap}} \quad (2)$$

where b_{sg} = scattering coefficient for gases (Rayleigh)

b_{sp} = scattering coefficient for particles

b_{ag} = absorption coefficient for gases

b_{ap} = absorption coefficient for particles

Particles of the fine fraction are the most effective in light scattering and of all airborne particles have the major effect on reducing visibility (2). In apportioning the contributions of the

fine particulate species to the extinction of light, sulfate scattering has been estimated to account for as much as 76% (3). In fact, Hasan and Dzubay (7) reported that differences in the scattering-to-mass-concentration ratios for all the major species in the aerosol were statistically insignificant, except for ammonium sulfate.

D. Measurement of Visual Range

Visual range, V , is the distance at which the apparent contrast between an object and its background becomes equal to the contrast threshold in the eyes of an observer (12). The extinction coefficient also relates to the apparent contrast of distant objects and Koschmieder showed in 1924 that

$$b_{\text{ext}} V = 3.9 \quad (3)$$

when the distant object is black and the observer's contrast threshold for detection is 2%. The significance of Equation (3) is the connection provided between an instrumentally-measured light extinction and a visual range directly associated with human perception of air pollution (6).

The standard method for determining visibility is human observation (2). However, various instrumental techniques have been developed to measure visual range so that transmissometers, integrating nephelometers, telephotometers, and photographic techniques are used. Of these, it appears that the telephotometer and the camera best approximate the visibility perception of the eye (13).

For this reason, a photographic technique recommended by EPA-RTP was employed in the present study to accompany human observation.

CHAPTER II

EXPERIMENTAL PROCEDURE

A. Visual Range Measurements

Measurements were made at noon on aerosol sampling days from atop Howard's Knob, located approximately 2 km northwest of downtown Boone. Visibility photographs were taken of the same southwestern view of mountain ranges with a 35 mm Vivitar 220/SL single-lens reflex camera, using ASA 64 Kodak Ektachrome color slide film. Three different exposure settings were used for each observation. Visibility was estimated by the observer and noted at the time the photographs were taken.

Projected slides were used to identify the most distant mountain peak having distinguishable contrast with the background. The identity of the peak and its distance were determined from U. S. Geological Survey topographical maps so that the distance from Howard's Knob to that peak was taken as the visual range for the sampling day.

B. Aerosol Measurements

Measurements were made from a rural area characterized by pasture and farmland approximately 5 km southeast of downtown Boone. The site was a 40 X 25 m clearing in the forest about 20 m from a small river. Bounding the site on two sides was a sparsely

traveled dirt road, and on a third side was a one-family residence from which electrical power was obtained.

From the center of this site aerosol samples were collected by the operation of a Beckman Automatic Dichotomous Sampler (Figure 1). Ambient air was drawn through the sampler and the particulate matter separated into fine and coarse fractions by a virtual impactor (Figure 2), with the separation point specified as 2.5 μm diameter. These fractions were collected on pairs of Teflon filters obtained from Ghia Corporation, Pleasanton, California. Microprocessor control of the sampler allowed for programming 24 hour sampling periods, midnight to midnight, every third of fourth day throughout the study period. Flow rate at the sampler inlet was measured following the study with a GCA/Precision Scientific Wet Test Meter, indicating that a 16.7 L/min flow rate ($1 \text{ m}^3/\text{hr}$) was maintained from initial calibration to the end of the study.

Midway and at the conclusion of the study period, filter trays containing sample pairs were removed from the dichotomous sampler for analysis. Both sets of filters were analyzed for sulfate content using a Dionex Ion Chromatograph Model 10 (Figure 3). The method consisted of ion-exchange liquid chromatography of the filter extract with eluent suppression and conductimetric detection (16). The total fine particle mass on each of the second set of filters was determined by the process of beta-gauging. This method involved measuring the intensity of an electron beam (^{147}Pm)

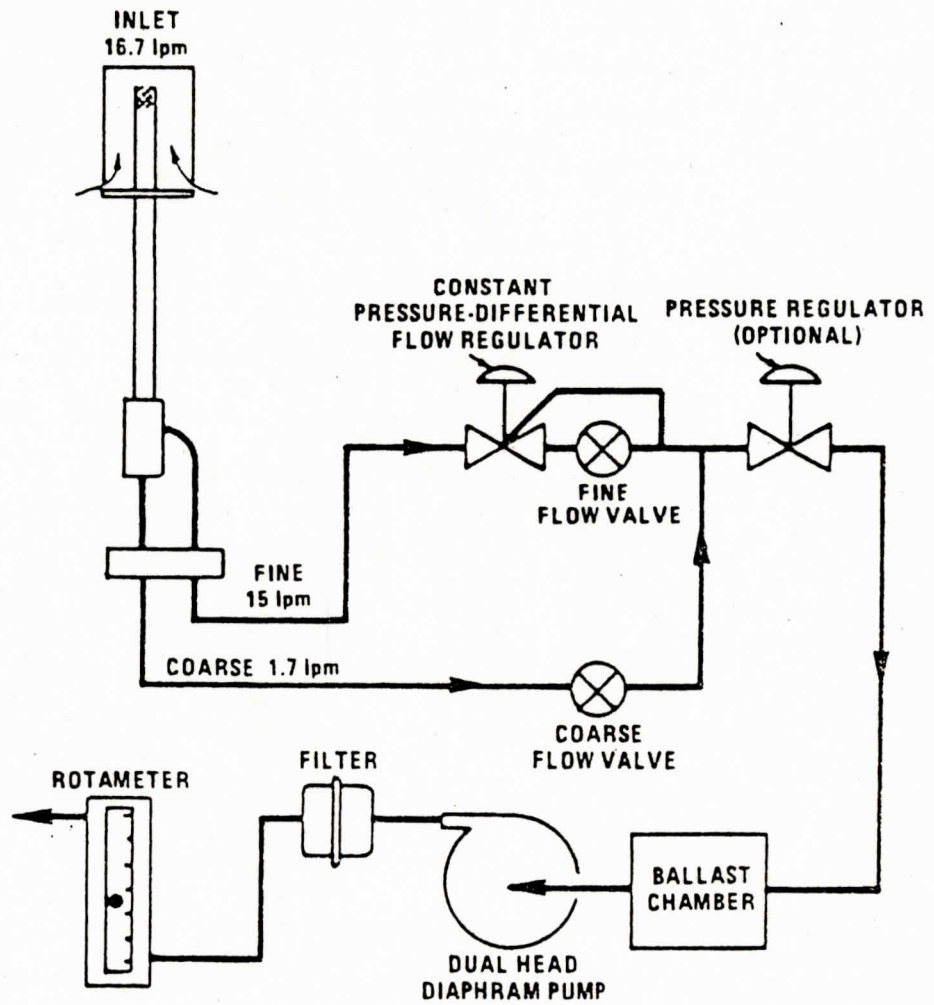


Figure 1. Schematic view of the Beckman dichotomous sampler (14).

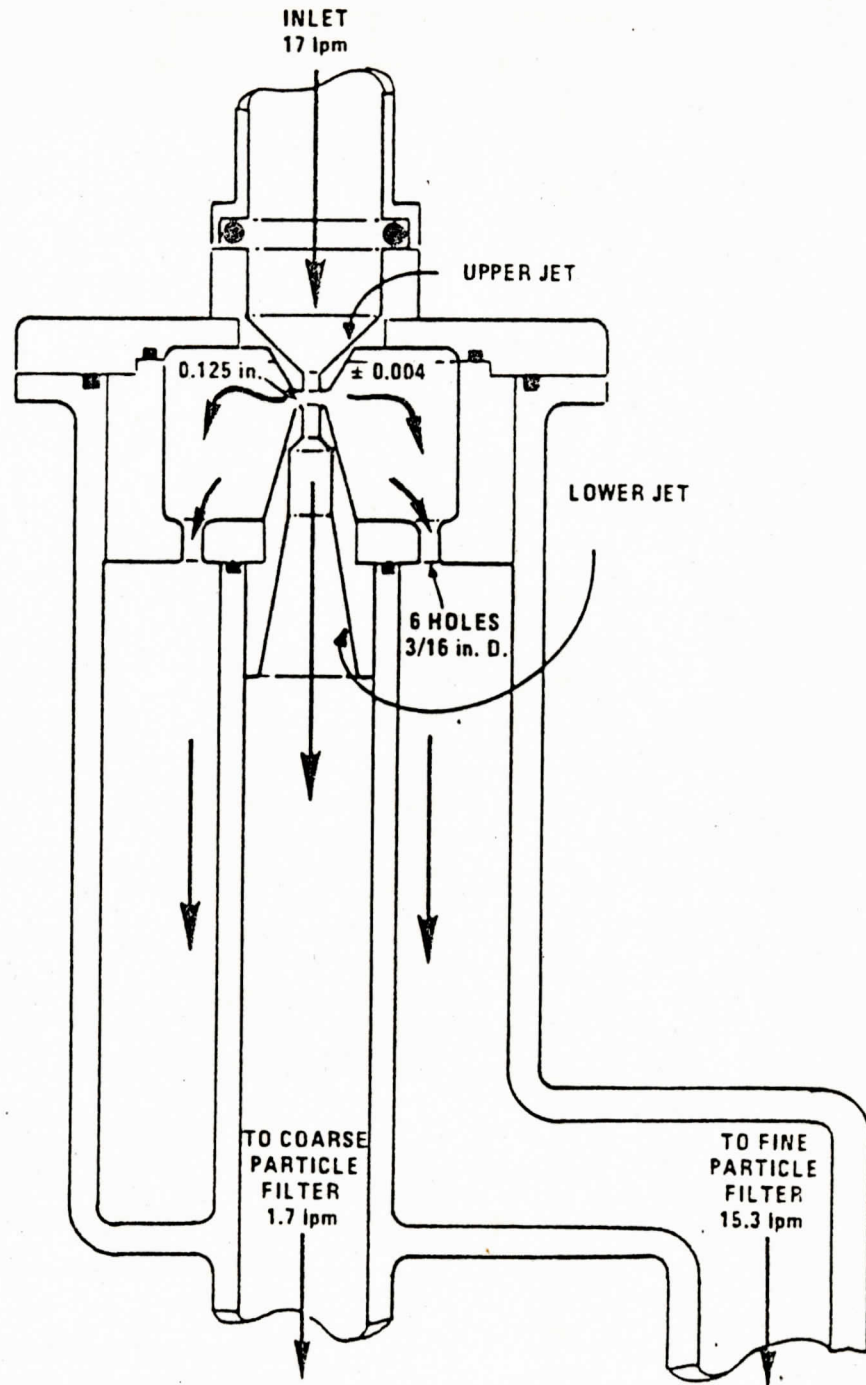


Figure 2. Cross-sectional view of virtual impactor designed by Loo et al., 1978 (14).

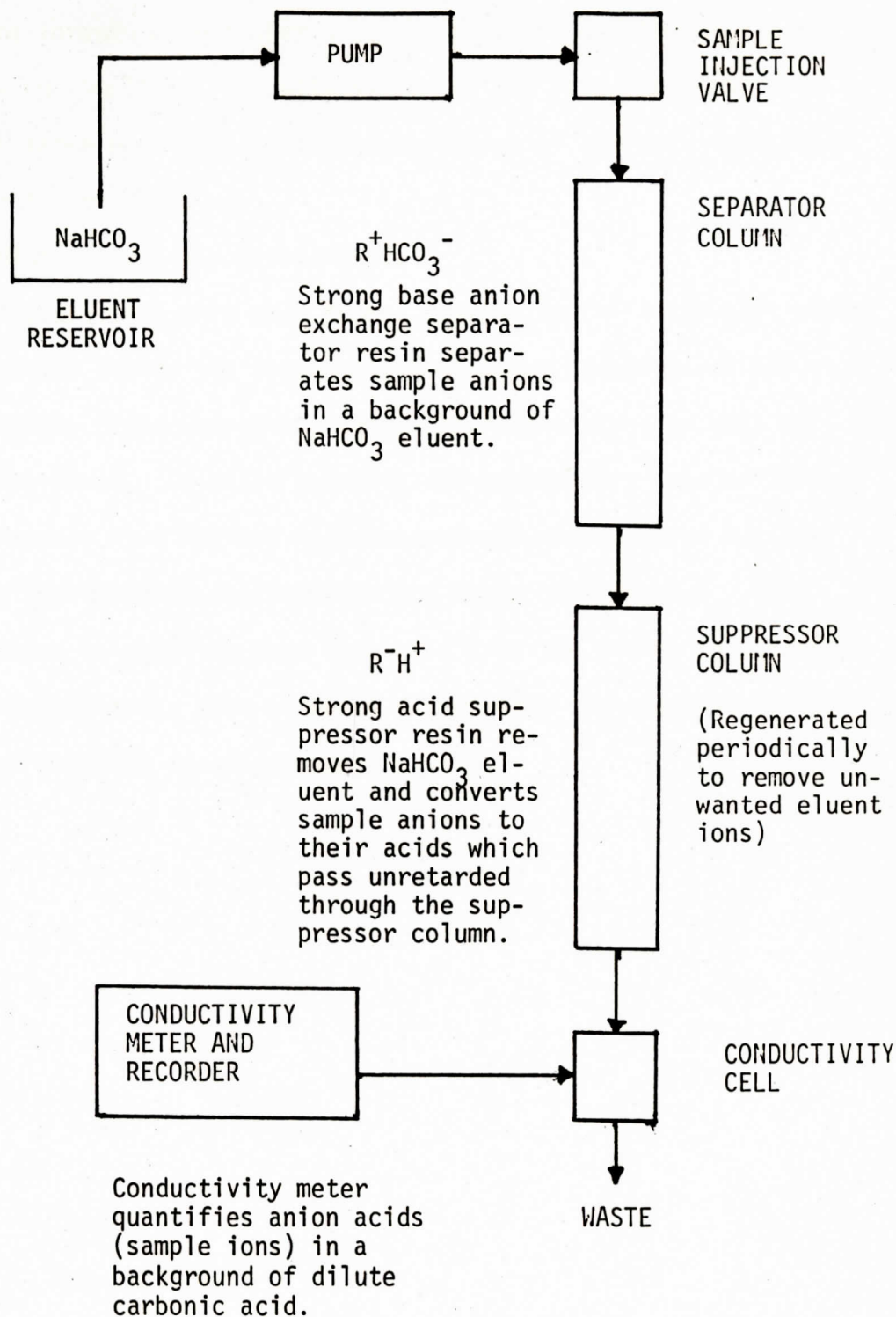


Figure 3. Dionex ion chromatograph flow scheme illustrating anion analysis (15).

passing through each filter prior to sampling and then again following sampling. The attenuation of the beta radiation due to the laden particles enabled computation of the mass deposited on the filter (17).

C. Relative Humidity Measurements

Measurements were made from a meteorological instrument shelter located behind the Center for Continuing Education on the Appalachian State University campus approximately 1 km west of downtown Boone. Relative humidity and temperature were monitored continuously by a Belfort Hygrothermograph which sensed humidity by the expansion or contraction of a human hair element. Noon humidities on sampling days were read from the charts to accompany noon visibilities, both of which represented the midpoint of the aerosol sampling duration time.

CHAPTER III

RESULTS AND DISCUSSION

The ion chromatographic analysis of laden filters collected by the dichotomous sampler yielded the sulfate content of the fine particle fraction. Assuming acid sulfate neutralization from ambient bases prior to analysis, sulfate levels were calculated as ammonium sulfate. Levels were determined as the average concentrations in the ambient air during the 24 hour sampling period, calculated from total sulfate mass and flow rate through the dichotomous sampler. The master data table (Table I) indicates the fine particulate sulfate level for each sampling day with the corresponding total fine particle mass, visual range, and relative humidity measurements. The supplemental data table (Table II) includes those samples from the first set of filters for which the total fine particle mass was not determined or which were not synchronized with visual range measurements due to dichotomous sampler programming difficulties.

Figure 4 shows the correlation between fine particulate sulfate mass concentration (M_s) and visual range (V). Relative humidity (RH) is indicated beside each data point. Two points were excluded from calculation of the correlation coefficient, based on relative humidity factors. Ferman et al. (3) have shown that ambient aerosol light extinction due to scattering by associated water

TABLE I. MASTER DATA TABLE

Sampling Date	Fine Particulate Sulfate ($\mu\text{g m}^{-3}$)*	Fine Particulate Mass ($\mu\text{g m}^{-3}$)**	Noon Relative Humidity (%)	Noon Visual Range (km)
7-25-81	16.6	-	70	10
8-9-81	11.6	-	70	20
9-2-81	5.04	-	75	6
10-18-81	6.19	13	-	-
10-22-81	3.71	8.0	47	39
11-4-81	5.92	16	46	20
12-3-81	1.39	5.6	50	-
12-7-81	2.86	7.2	76	39
12-11-81	2.70	6.1	56	39
12-15-81	2.17	6.9	-	-
12-19-81	4.11	8.5	59	-
12-23-81	2.94	5.3	-	-

*Calculated as ammonium sulfate

**Filter deposit area is 6.4 cm^2

TABLE I. MASTER DATA TABLE (CONTINUED)

Sampling Date	Fine Particulate Sulfate ($\mu\text{g m}^{-3}$)*	Fine Particulate Mass ($\mu\text{g m}^{-3}$)**	Noon Relative Humidity (%)	Noon Visual Range (km)
1-8-82	2.02	6.7	79	72
1-12-82	3.62	10	88	-
1-16-82	3.69	13	-	-
1-20-82	7.57	13	50	20
1-24-82	2.20	6.4	47	-
1-28-82	6.63	18	81	-
2-1-82	4.46	9.9	57	72
2-16-82	0.48	9.3	93	-
2-22-82	6.27	10	70	-
2-24-82	2.52	11	43	39
2-26-82	2.85	9.1	90	10

*Calculated as ammonium sulfate **Filter deposit area in 6.4 cm^2

TABLE II. SUPPLEMENTAL DATA TABLE

Sampling Date	Fine Particulate Sulfate ($\mu\text{g m}^{-3}$)*	Noon Relative Humidity (%)
7-28	8.20	80
7-31	5.20	73
8-3	12.5	90
8-6	11.8	95
8-12	13.5	78
8-18	4.68	60
8-21	1.87	54
8-24	30.1	85
8-27	21.8	64
8-30	6.77	72
9-11	9.69	58
9-14	12.8	78
9-20	2.70	55
9-23	4.35	46
9-26	6.59	47
9-29	2.70	70
10-2	2.31	60
10-5	5.28	92
10-9	3.81	65
10-13	5.10	61

*Calculated as ammonium sulfate

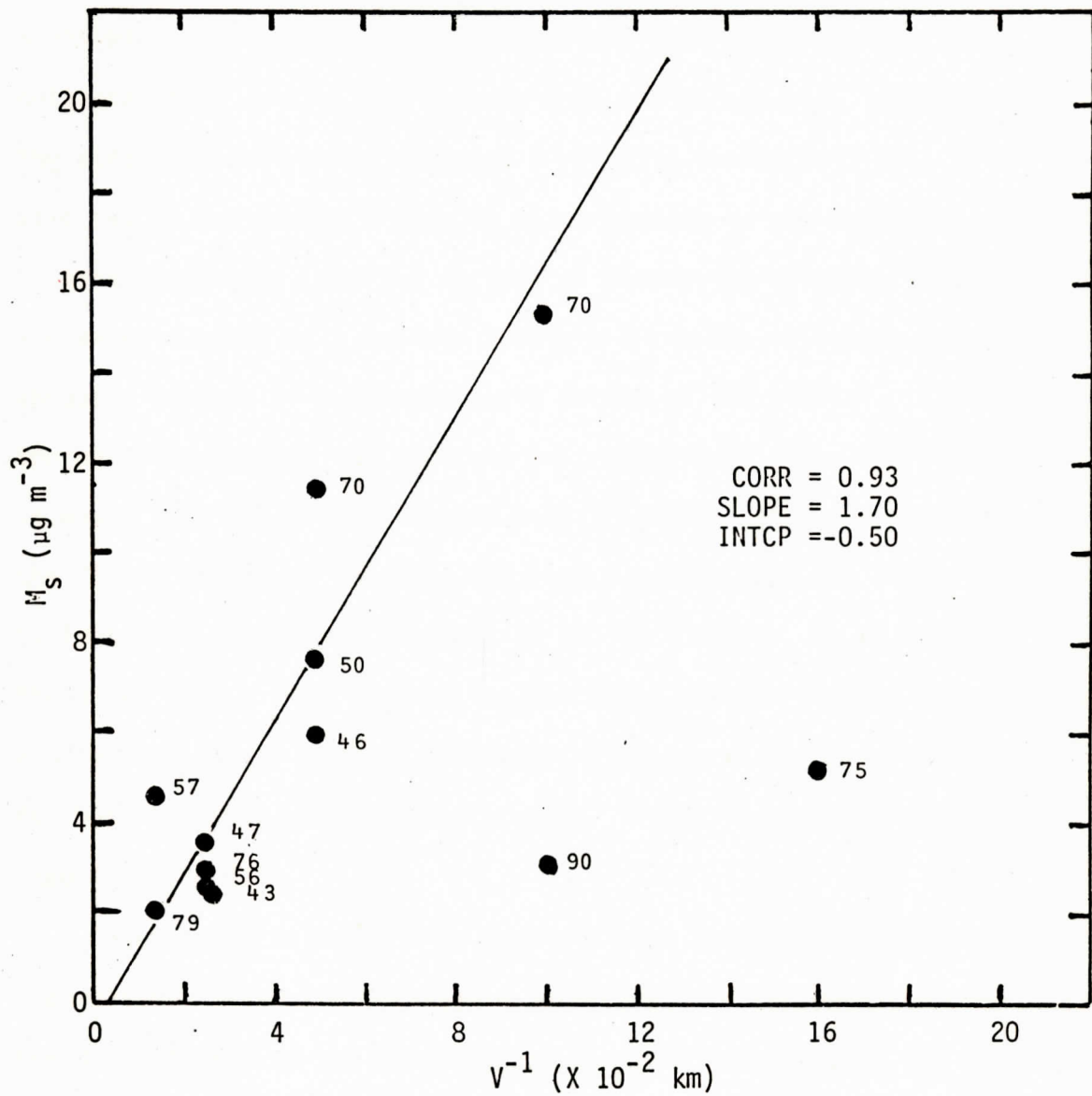


Figure 4. Fine particulate sulfate concentration vs. inverse visual range, with relative humidity indicated beside each data point.

molecules is disproportionately large for relative humidity greater than about 80%. Therefore, as expected, the 90% RH point exhibited a visual range that was far less than on lower humidity days when collected fine particulate sulfate mass was comparable. Although the other excluded point reported a relative humidity of 75%, it also may be considered a high RH point because of the phenomenon of hysteresis (18). During periods of decreasing relative humidity, the water content of a sulfate particle decreases, although not abruptly because of the hygroscopic nature of the aerosol. Thus the water content of the particle may remain well above the value it would have had if equilibrium were attained. It was noted that the 75% RH point was taken during such a period when the relative humidity was decreasing from 90% to 70% in 4 hours. By rejecting these two data a high correlation was found between sulfate levels and reduced visual range. Similarly, high correlations were found for both the Shenandoah Valley studies. These results are summarized in Table III.

Calculation of total fine particle mass concentration (M_f) was accomplished using the beta-gauge determinations for particle mass, the deposit area on the filters, and the flow rate through the dichotomous sampler. The sulfate percentage contribution to the total fine particle fraction was computed for each sampling day, and the average percent sulfate level for the study period is presented in Table III along with the values from several other aerosol studies. The mean sulfate level for Boone was found to be within the lower portion of the range of reported values.

TABLE III. SUMMARY OF RECENT AEROSOL STUDIES

Study	Corr. M_s, V^{-1}	Aerosol Sulf. (%)	$V \cdot M_f^2$ (gm^{-2})
Ferman et al., (3) Shenandoah Valley, VA July and August, 1980	0.91 ^a	55	0.73 ^a
Stevens et al., (5) Shenandoah Valley, VA July 17-August 5, 1980	≈0.87	-	0.46
Stevens et al., (4) Great Smoky Mountains, TN September 20-26, 1978	-	61	-
Lodge et al., (2) literature review, 1980	-	-	1.25 ^b
Dzubay et al., (6) Houston, TX September 11-19, 1980	-	54	0.30±0.04 0.28±0.04 ^c
Trijonis, (9) 8 sites, Rural East Annual and Summer, 1976-80	-	31	-
Rao and Sistla, (8) Niagara Frontier, NY September 1978-March 1979	-	25	-
Wilson, present study Northwestern NC July 1981-February 1982	0.93	38	0.19 0.27 ^d

^a determined from b_{sp} measurements

^c empirical derivation

^b theoretical derivation

^d corrected value

Figure 5 indicates the correlation between total fine particle mass concentration and visual range. Again the high relative humidity data point was excluded from calculation of the correlation coefficient. Although the correlation is not exceptionally good, it must be remembered that the sampling period spanned both hot, humid summer weather and very cold winter weather. It is possible that the composition of the fine particle mass was not constant over this period.

The slope of Figure 5 yields a best fit value of 0.19 g m^{-2} for the $V M_f$ product during the study period. Several recent investigators (2-6) believe this product to be a constant, assuming that light extinction is proportional to the total fine particle mass;

$$b_{\text{ext}} = k M_f \quad , \quad (4)$$

so that substitution into Equation (3) gives

$$V M_f = K \quad . \quad (5)$$

The value for this constant has been the subject of recent controversy. The $V M_f$ product values obtained for several studies are summarized in Table III for comparison.

Recalling from Equation (2) that Rayleigh scattering contributes to the extinction coefficient, it is apparent that this becomes a significant factor for atmospheric light extinction when particle concentrations are low and visibility is long range. Possibly this accounts for some fluctuation in the $V M_f$ product for this study. Dzubay (19) has suggested inserting a "mass-equivalent" correction for Rayleigh scattering so that Equation (5)

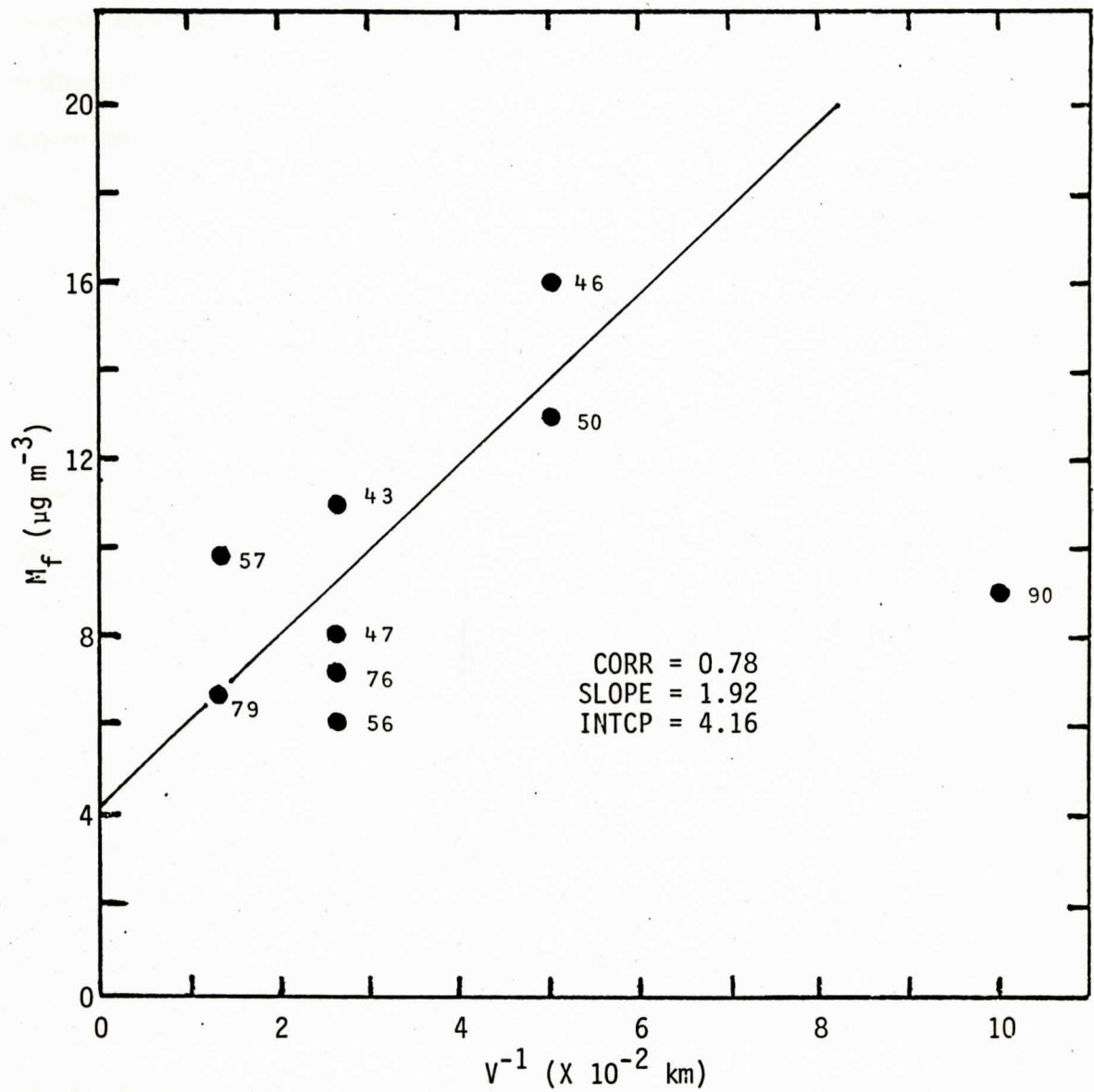


Figure 5. Total fine particle mass concentration vs. inverse visual range, with relative humidity indicated beside each data point.

may be rewritten as

$$V \left[M_f + M_f (\text{Rayleigh}) \right] = K \quad (6)$$

Dzubay estimates the correction term to be $-3 \mu\text{g m}^{-3}$, based on a meteorological range constant of 2.9 (5.5% contrast threshold), a Rayleigh scattering coefficient of 10.5 Mm^{-1} , and a specific scattering efficiency of $3.5 \text{ m}^2 \text{ g}^{-1}$. When this estimated mass-equivalent correction is applied to all points of Figure 6, the slope is of course unchanged at 0.19 g m^{-2} , but the infinite V extrapolation (y-intercept) is 1.2 g m^{-3} .

In addition to the Rayleigh scattering effect, there exist inherent uncertainties in determining $V M_f$ at long visual range, including:

- (1) point-in-time V determinations versus 24 hour integrated M_f ,
- (2) large relative mass measurement uncertainties at low mass (20),
- (3) observer error in estimating V over long distances through inhomogeneous atmospheres (21).

There is therefore some justification for excluding long V data. If the aforementioned corrected data is restricted to visual ranges of 39 km or less, the slope of the resulting best fit line is 0.27 g m^{-2} with an intercept of $1.1 \mu\text{g m}^{-3}$. Although the data is limited, it is interesting that elimination of long-range data serves to correct the $V M_f$ product value of this study to compare extremely well with the empirical value derived by Dzubay et al. (6).

Since the majority of data obtained during the study period was for fine particulate sulfate levels, a bar graph of the total sulfate data base by weekly average is presented in Figure 6. The graph also indicates total fine particle mass level averages where they were known. It was found that for this study period there was a trend toward lower sulfate concentrations as summer progressed to winter. The following are the computed seasonal M_s means in $\mu\text{g m}^{-3}$:

summer (July 25 → September 22) 10.9 ± 7.4 (16 points)

fall (September 23 → December 21) 4.0 ± 1.6 (15 points)

winter (December 22 → February 26) 3.8 ± 2.1 (12 points).

Upon removal of the extremely high sulfate points on August 24 and 27 (due to local meteorological stagnation), the summer average becomes $8.8 \pm 4.5 \mu\text{g m}^{-3}$. These averages are consistent with Husar et al. (22), who found increased summer sulfate concentrations to correspond with increased use of coal in this geographical region during the summer months.

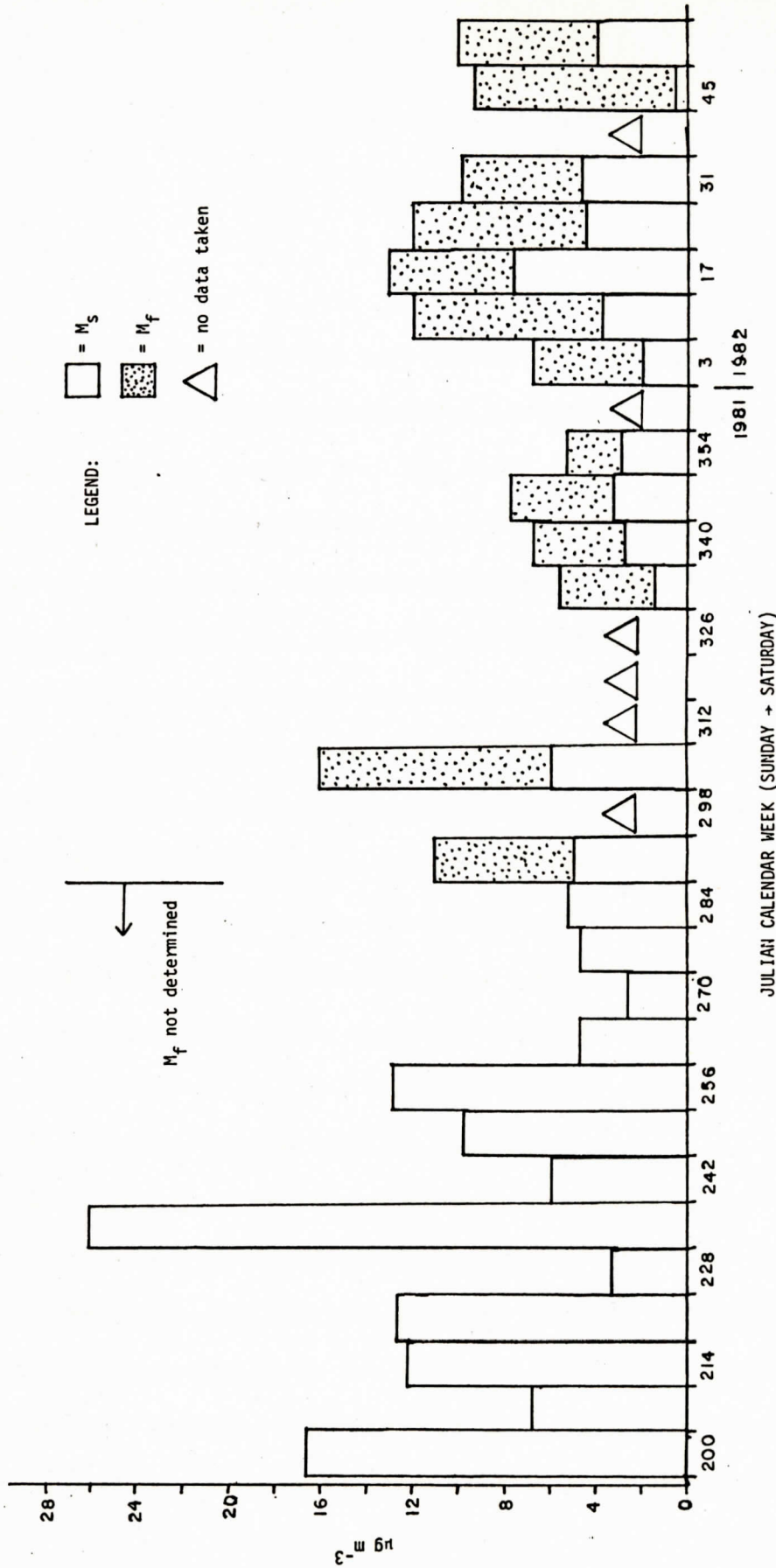


Figure 6. Weekly average bar graph for fine particulate sulfate and total fine particle mass concentrations through the study period.

CHAPTER IV

CONCLUSION

It was found that a high correlation exists between fine particulate sulfate mass concentration and reduced visibility. Relative humidities greater than 80% acted to further reduce visual range. These results concurred with those of similar studies in the southern Appalachian mountains. However, the sulfate percentage of the total fine particle mass, while within the reported range of values, was somewhat lower than the sulfate levels of the Shenandoah Valley and the Great Smoky Mountains. As expected, the average seasonal sulfate levels during the seven month study period exhibited a decreasing trend from summer to winter. Although there was a lower correlation between total fine particle mass and reduced visual range, a corrected value for the $V M_f$ product from this study compared well with an empirical value derived by Dzubay et al., and was significantly different from products obtained in other similar studies.

Two improvements to this experiment are suggested for future investigations: (1) visual range measurements should be consistently coordinated with aerosol sampling, possibly with multiple determinations to obtain an average daily visibility; and (2) meteorological observations should be more closely coordinated with the data obtained. Variations on this experiment could

concentrate on expansion of the data base to include additional analytical information relevant to aerosol chemistry in this geographical region. Determinations of levels of elemental and organic carbon, nitrogen oxides, and other species present in the fine aerosol would better characterize the nature of the ambient air in the mountains of northwestern North Carolina.

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