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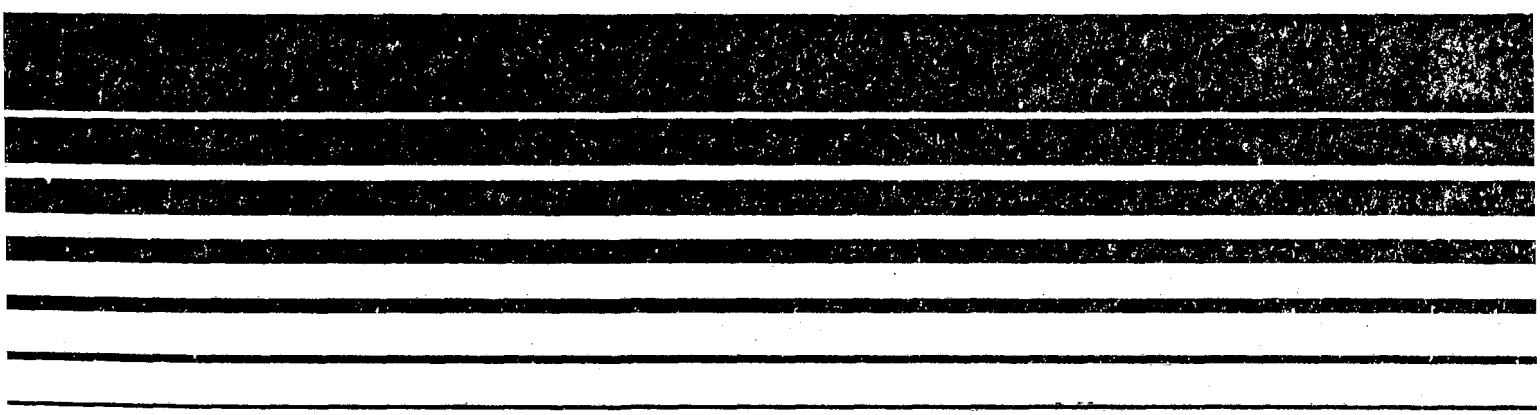

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Existing and Natural Background Levels of Visibility and Fine Particles in the Rural East

by

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ABSTRACT

An investigation is conducted into existing and natural background levels of visibility and fine particles in nonurban areas of the Eastern U.S. An analysis of data for 100 airports nationwide indicates that nonurban areas of the East experience relatively low visibilities; specifically, rural areas east of the Mississippi and south of the Great Lakes generally exhibit annual median visual ranges of 10-15 miles. A review of data from eight monitoring programs indicates that ambient fine ($\leq$ 2.5 μm) particle concentrations presently average approximately 29 $\mu g/m^3$ in the rural East. The largest components are water ($\sim$ 11 $\mu g/m^3$), sulfates ($\sim$ 9 $\mu g/m^3$), and organics ($\sim$ 4 $\mu g/m^3$). The majority of the water is thought to be attached to hygroscopic sulfate aerosols. Currently, sulfates and fine particles exhibit a pronounced maximum in the summer quarter, when visibility shows a pronounced minimum. This seasonal pattern is a new phenomenon historically; prior to the 1960s, visibility was distinctly higher during the summer than during the remainder of the year. An investigation of natural background conditions suggests that natural fine aerosol concentrations would average $5\frac{1}{2} \pm 2\frac{1}{2}$ $\mu g/m^3$ in the East, with the largest components being organics and water. Natural background visual range for the East is estimated to be 60 $\pm$ 30 miles. It is not currently possible to check this estimate of natural background visual range through an analysis of historical visibility trends from the 1930s to the 1970s because of limitations in historical emission trend data, uncertainties in airport visibility trend data, and the confounding effects of meteorology. The best check would be to collect simultaneous measurements of fine particle mass, fine particle chemical composition, and visibility in remote continental areas of the Southern Hemisphere.

INTRODUCTION

During the past decade, numerous studies have been conducted of large-scale regional air quality problems in the Eastern United States. Based on the results of these studies, it is now generally accepted that the Eastern U.S. experiences a regional problem with respect to fine aerosols (particles $\lesssim$ 2.5 μm in diameter). Fine aerosol concentrations in rural areas of the East significantly exceed those in other areas of North America, and visibility in the rural East is correspondingly low. Because acid sulfate particles constitute a large fraction of the Eastern aerosol (Pierson et al. 1980; Stevens et al. 1980; Ferman et al. 1981), the fine aerosol and visibility problems may be closely linked with the regional acid rain phenomenon.

One major purpose of this paper is to characterize existing levels of visibility and fine particles in rural areas east of the Mississippi. In this paper, we document existing visibility patterns by analyzing three years of visual range data from one hundred airports. We then construct a chemically-resolved model fine aerosol -- annually and spatially averaged for the nonurban East -- by analyzing recent particulate data from eight monitoring programs. Seasonal variations in visibility and fine particles are also considered.

The second major goal is to estimate natural background conditions, so that natural versus man-made contributions to existing fine particle concentrations can be better understood. A chemically-resolved model fine aerosol for natural background conditions is determined based on emission calculations, trace element calculations, and data collected in remote areas. The model natural aerosol yields an estimate of natural background visibility. We also investigate the possibility of checking this estimate of natural background

visibility through an analysis of historical visibility trends from the early 1930s to the early 1970s.

EXISTING VISIBILITY LEVELS

In order to characterize existing visibility levels in rural areas of the East, and in order to compare Eastern visibility to visibility elsewhere in the nation, we have conducted a comprehensive analysis of daytime visual range measurements at 100 suburban/nonurban airports nationwide (Trijonis and Shapland 1979). Daytime visual range measurements at airports are made by observing distant markers (e.g. mountains or buildings) against the horizon sky. Before selecting locations for this study, telephone surveys were conducted at each airport to insure that an adequate set of markers was available for estimating visual range. Specifically, we attempted to select only those airports that had very distant markers relative to the average visual range for the surrounding region. In a few cases, however, in order to attain complete geographical coverage, we were forced to use airports with markers at relatively short distances (e.g. at only 10 to 15 miles).

Our analysis of existing visibility levels is based on median visual ranges calculated from cumulative frequency distributions of the airport data. Because the nature of reporting practices at airports leads to an implicit "greater than or equal to" meaning in the visual range recordings, special techniques must be applied in determining these cumulative frequency distributions (Trijonis 1979; Trijonis and Yuan 1978; Trijonis and Shapland 1979; Husar et al. 1979; Latimer et al 1979; Sloane 1980). Specifically, the cumulative frequency distribution must be given in the form "percent of time visual range is greater than or equal to X miles", starting with the

most distant marker and proceeding sequentially to the less distant markers. Furthermore, the points in the cumulative frequency distribution should be plotted only at the visual ranges that are routinely reported by the airport observation team; otherwise artificial "kinks" will be produced in the frequency distributions. Application of these special techniques should make the distributions consistent from site to site, even if the airports have visibility markers at different distances.

Figure 1 presents examples of cumulative frequency distributions for six rural locations in various parts of the United States. The dots in Figure 1 represent the routinely reported visual ranges (often these correspond to distinct visibility markers). The lines drawn between the dots represent linear interpolations of the cumulative frequency distributions. As is the case with five of the six locations in Figure 1, most of the median visual ranges are determined by linear interpolations of the cumulative frequency distribution. At some sites (e.g. Madison WI in Figure 1), however, the frequency distributions require extrapolation beyond the farthest marker in order to reach the median visual ranges. The forms of these extrapolations, linear or nonlinear, are based on a comparison of the distribution for the station in question with distributions for other sites located in the same general geographical area.

Based on the above analytical procedure, we have derived Figure 2, a map of median mid-day visual ranges for rural areas throughout the United States. The rather high quality of the data in Figure 2 -- at least in terms of internal consistency -- is suggested by the monotonic gradients that often exist in passing from areas of poor visibility to areas of good visibility, and by the agreement of the readings among neighboring stations.

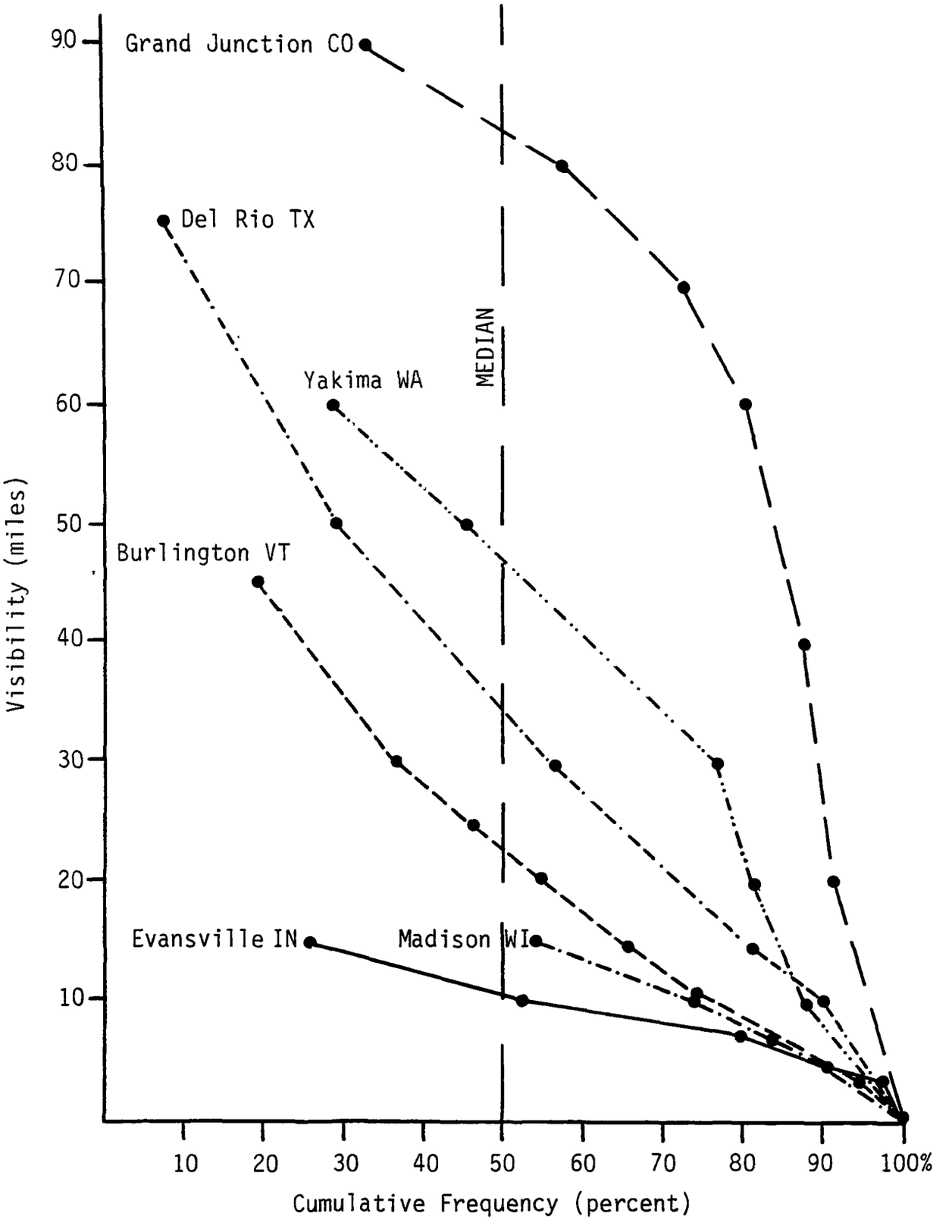

Figure 1. Examples of cumulative frequency distributions for visibility (data for mid-day hour, 1974-1976).

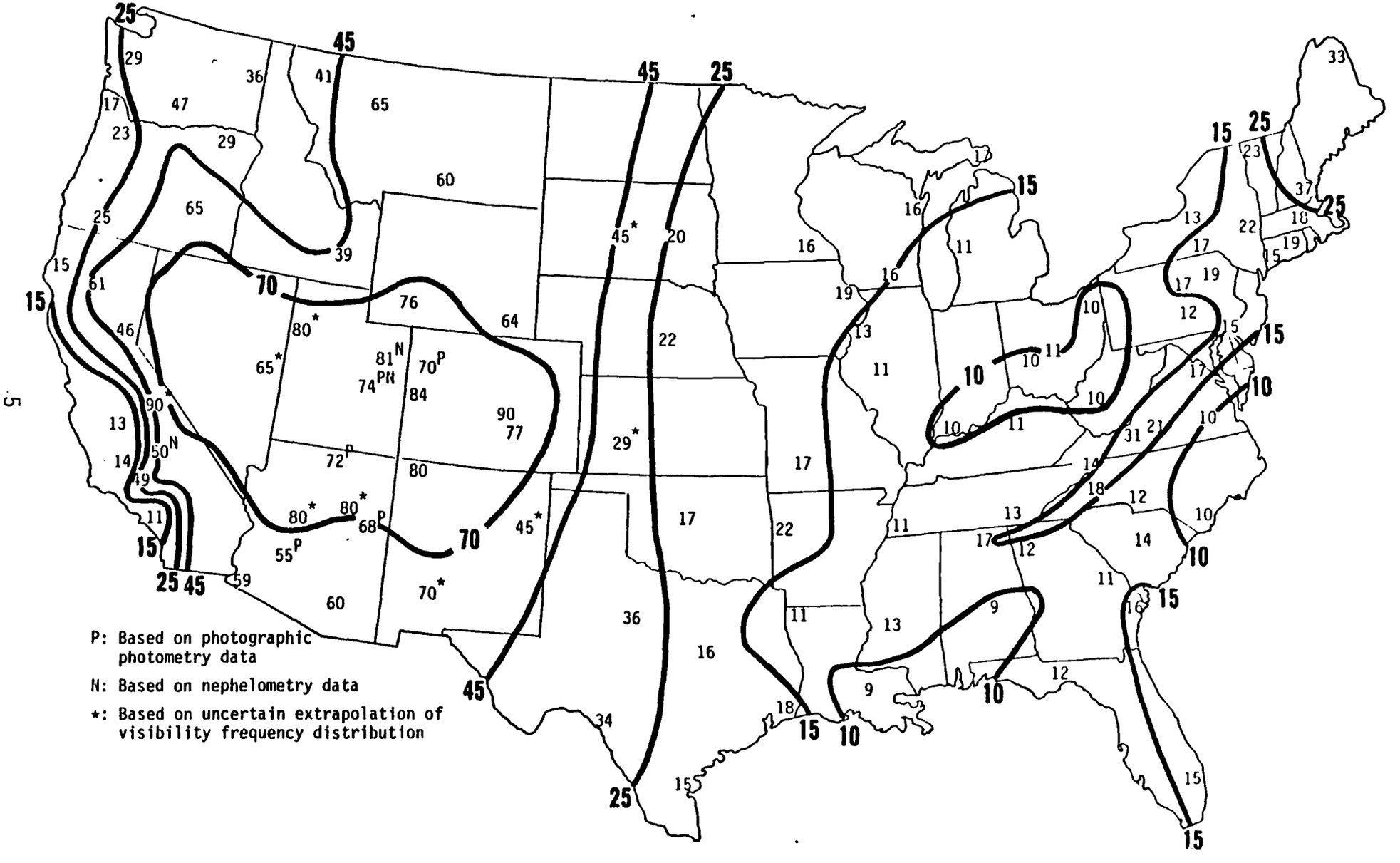

Figure 2. Median annual visual range (in miles) at suburban/nonurban locations in the United States (1974-1976).

Figure 2 demonstrates that the mountainous Southwest experiences the best visibility in the country. Specifically, median visual range exceeds 70 miles in a region comprised of Utah, Colorado, Nevada, northern Arizona, northwestern New Mexico, and southwestern Wyoming. Visual range is also quite good, exceeding 45 miles, to the north and south of this region. Passing westward or eastward, fairly sharp gradients occur. Median visual range falls to less than 25 miles in a narrow band along the northern Pacific coast and to less than 15 miles in the Central Valley of California and the Los Angeles basin. Although some parts of the East (e.g. northern New England) experience moderate visibility levels ($\sim$ 25 miles), median visual range is generally less than 15 miles in areas east of the Mississippi and south of the Great Lakes. This Eastern area of low visibility is the focus of the present paper.

EXISTING FINE PARTICLE CONCENTRATIONS

Table 1 presents average mass concentrations of fine particles ($\lesssim$ 2.5 μm) obtained from several monitoring programs in rural areas of the Eastern U.S. Data are presented not only for total fine particle mass but also for fine particle chemical composition. Because some of the monitoring programs operated only during the summer, Table 1 is divided according to summertime and annual monitoring programs.

As indicated in the left-hand column and the footnotes of Table 1, we have made several adjustments in order to establish a consistent basis for comparing data from the various monitoring programs. For example, we have reported all the sulfate mass data as ammonium bisulfate (NH_4HSO_4), because

TABLE 1. SUMMARY OF AVAILABLE DATA ON FINE PARTICLE CONCENTRATIONS IN RURAL AREAS OF THE EASTERN U.S.

	ANNUAL MONITORING PROGRAMS					SUMMERTIME MONITORING PROGRAMS		
STUDY	EPA Dichotomous Sampler Network	Dzubay (1980) Trijonis et al. (1980)	Watson et al. (1981) Mueller (1981)	EPA NASN Network	Daisey et al. (1979)	Stevens et al. (1980)	Ferman et al. (1981)	Pierson et al. (1980)
SAMPLING LOCATIONS	Average of Will Cnty. IL, Erie Cnty. NY, and Durham Cnty. NC	Average of two rural sites near St. Louis	Average of nine nonurban SURE sites	Average of twenty nonurban Eastern sites	Sterling Forest, New York State	Great Smokey Mts. in Tennessee	Luray, VA	Allegheny Mt. in Pennsylvania
SAMPLING PERIOD	One year, 10/79 to 9/80	One year, 1976	Six months of data 8/77-10/78	Three years, 1974 to 1976	Two years, 1977 to 1978	Six days in September 1978	One month in the summer of 1980	Eighteen days in the summer of 1977
AVERAGE FINE AEROSOL MASS [$\mu g/m^3$]								
Sulfates (as NH_4HSO_4)	7.7	8.1	7.7	9.2*	NA	13.5	13.3	14.3
Organics (as $CH_2O_{1.4}$)	NA	NA	NA	NA	2.8	3.3	7.1	NA
Elemental Carbon	NA	NA	NA	NA	NA	1.1	1.3	<0.8
Crustal (as 4 x Si)	1.4	1.4	NA	NA	NA	0.2	2.0	NA
Nitrate (as NH_4NO_3)	1.0	NA	NA	NA	NA	0.3	0.2	<0.7
Other (Water?)	NA	NA	NA	NA	NA	5.6	0.0	NA
TOTAL	20.3	16.5	19.0	NA	NA	24.0	23.9	NA

NA: Not available

* The NASN data are for total particulate sulfate: a factor of 0.9 has been applied to account for size distribution effects and to yield an estimate of fine sulfate. Also, note that, unlike the other sulfate data in this table which are from teflon filters, the NASN sulfate data are subject to overestimation due to artifact sulfate formation on glass fiber filters.

several investigators have found that the average cation associated with $SO_4^=$ in the East corresponds approximately to $(NH_4H)^{++}$ (Pierson et al. 1980; Stevens et al. 1980; Ferman et al. 1981). Also, following Duce (1978), Hahn (1980), and Chu and Macias (1981), we have assumed that the conversion factor from organic carbon aerosol to total organic aerosol is 1.5 (corresponding to an average composition of $CH_2O_{\frac{1}{4}}$).

The mass concentration and chemical composition of fine particles in the East exhibits significant variations both seasonally and spatially. For the purposes of the discussions in this and subsequent sections, we would like to postulate a single "model" ambient aerosol representing annual mean concentrations spatially averaged over rural Eastern areas. Table 2 presents this model ambient aerosol, which is based on the data in Table 1 and the following considerations: The summertime sulfate averages in Table 1 should be divided by approximately 1.3 to make them comparable to annual averages; data for 20 rural EPA NASN sites and 3 rural EPA Dichotomous Network sites in the East indicate that summertime (3rd quarter) sulfate averages are generally 1.1 to 1.6 times greater than annual sulfate averages. The summertime nitrate averages in Table 1 are likely to be low compared to annual nitrate averages; data from the EPA Dichotomous Network suggest that winter nitrate levels are nearly an order of magnitude greater than summer nitrate levels. One might a priori expect (and a cursory examination of Table 1 suggests) that summer organic levels in rural areas are higher than annual organic levels. However, data collected by Daisey et al. (1979) reveal no seasonal pattern in organic aerosol concentrations. We have not explicitly discounted for seasonal variations in using the organic data in Table 1 to arrive at our organic component in Table 2.

TABLE 2. MODEL AMBIENT FINE AEROSOL, ANNUAL MEAN FOR RURAL AREAS OF THE EAST.

COMPONENT	MASS CONTRIBUTION* ($\mu g/m^3$)
Sulfate (as NH_4HSO_4)	9 ± 2
Organics	4 ± 2
Elemental Carbon	1 ± ½
Crustal Material	1 ± ½
Nitrates (as NH_4NO_3)	1 ± ½
Non-water Other	2 ± 2
Water	11 ± 5
TOTAL	29 ± 6**

*Mass of particles less than 2.5 microns in size under ambient conditions (RH ∿ 70-75%).

**Total error is calculated as the root-mean-square of the individual errors.

The most important new consideration in constructing Table 2 involves the treatment of water. The aerosol samples in Table 1 represent filtered particulate matter weighed at 40 to 50% relative humidity; these samples likely contain substantially less water than does the ambient aerosol (Hidy et al. 1974; Tierney and Conner 1967; Demuynck 1975; Pierson et al. 1980). Unfortunately, definitive data are not available regarding mass concentrations of water in ambient aerosols at Eastern locations. We have calculated the mass of water associated with the fine rural Eastern aerosol using two methods. First, thermodynamic calculations (Tang 1981) as well as measurements made with microwave waterometers, nephelometers, and multi-stage cascade impactors (Hidy et al. 1974; Ho et al. 1974; Stelson and Seinfeld 1981; Covert et al. 1972, Countess et al. 1981) indicate that, at average Eastern humidities (70-75%), the mass of water associated with fine aerosols is approximately equal to, or slightly greater than, the mass of aerosol electrolytes (e.g. sulfates and nitrates). Noting that our model rural aerosol contains 10 $\mu g/m^3$ of sulfates and nitrates, the above referenced studies suggest that there should be 10 to 15 $\mu g/m^3$ of water in the fine aerosol. Second, we have calculated the water component using recent data taken at Luray VA (Ferman et al. 1981) and the following assumptions: (1) the mass concentrations of the non-water aerosol components are as listed in Table 2; (2) on the average, the ambient (wet) aerosol scatters 1.9 times as much light as the dry aerosol (Ferman et al. 1981); (3) the scattering per unit mass by dry sulfate is 1.5 times the scattering per unit mass by the remainder of the dry aerosol (Ferman 1981; Wolff et al. 1981); and (4) the scattering per unit mass by water is 1.7 times the scattering per unit mass by dry sulfate due to density differences (White 1918). A simple algebraic equation based on these data/assumptions can be

solved to yield a fine mass concentration for water of 8 $\mu g/m^3$. Considering the results of both methodologies, we have chosen 11 $\mu g/m^3$ as the water component in the fine rural Eastern aerosol.

Table 2 contains error bounds for each component in the annual mean, spatially averaged, model aerosol. These error bounds are based on our subjective evaluation of the uncertainties. For example, the error bounds for sulfate are relatively low, ± 20%, because the various monitoring programs all agree rather closely regarding fine sulfate concentrations (once adjustments have been made for seasonality). The error bounds for organics and water, on the other hand, are relatively high, ± 50%, because definitive data are not available for the organic and water components of the fine rural Eastern aerosol.

According to our model aerosol, the annual/spatial average fine particle concentration in rural areas of the East is 29 $\mu g/m^3$. The two largest constituents are water (11 $\mu g/m^3$) and sulfate (9 $\mu g/m^3$). Actually, because we expect that most of the water is attached to hygroscopic sulfate aerosols, these two constituents might almost be interpreted as a single predominant component that accounts for more than two-thirds of the fine aerosol mass. Such an interpretation would be consistent with various statistical studies indicating that visibility reduction in the East is tied very closely with high sulfate concentrations (Trijonis and Yuan 1978; Leaderer and Stolwijk 1979; Pierson et al. 1980; Ferman et al. 1981). The only other component that represents a substantial fraction (e.g. greater than 10%) of fine particle mass is organic aerosol, which constitutes about 4 $\mu g/m^3$.

SEASONAL PATTERNS IN VISIBILITY AND FINE AEROSOLS

Fine aerosol concentrations and visibility in rural areas of the East exhibit very strong seasonal variations. Figure 3 illustrates average seasonal patterns in rural Eastern areas for light extinction (proportional to the reciprocal of visibility), sulfate concentrations, and fine particle concentrations. All three parameters demonstrate a strong peak during the summer quarter.

The summertime maximum in fine particle and extinction levels in the East manifests itself not only in averages of data over many sites but also in data for almost any individual site; that is -- the summer peak in fine particles is nearly universal among rural Eastern locations. This phenomenon is illustrated by a visibility map prepared using 3rd quarter data (see Figure 4). Comparing Figure 4 to Figure 2, we see that the large area east of the Mississippi and south of the Great Lakes, which has a 10 to 15 mile median visual range annually, exhibits a very homogeneous 8 to 10 miles median visual range during the summer.

ESTIMATE OF NATURAL BACKGROUND CONDITIONS

A reasonable method of investigating natural background visibility and fine particle concentrations in the East is to estimate the natural and anthropogenic fractions for each chemical component in our "model" ambient aerosol (Table 2). We derive these estimates below based on ambient data collected in remote areas, emission calculations, and trace element calculations. In the process of making these estimates, we held rather long

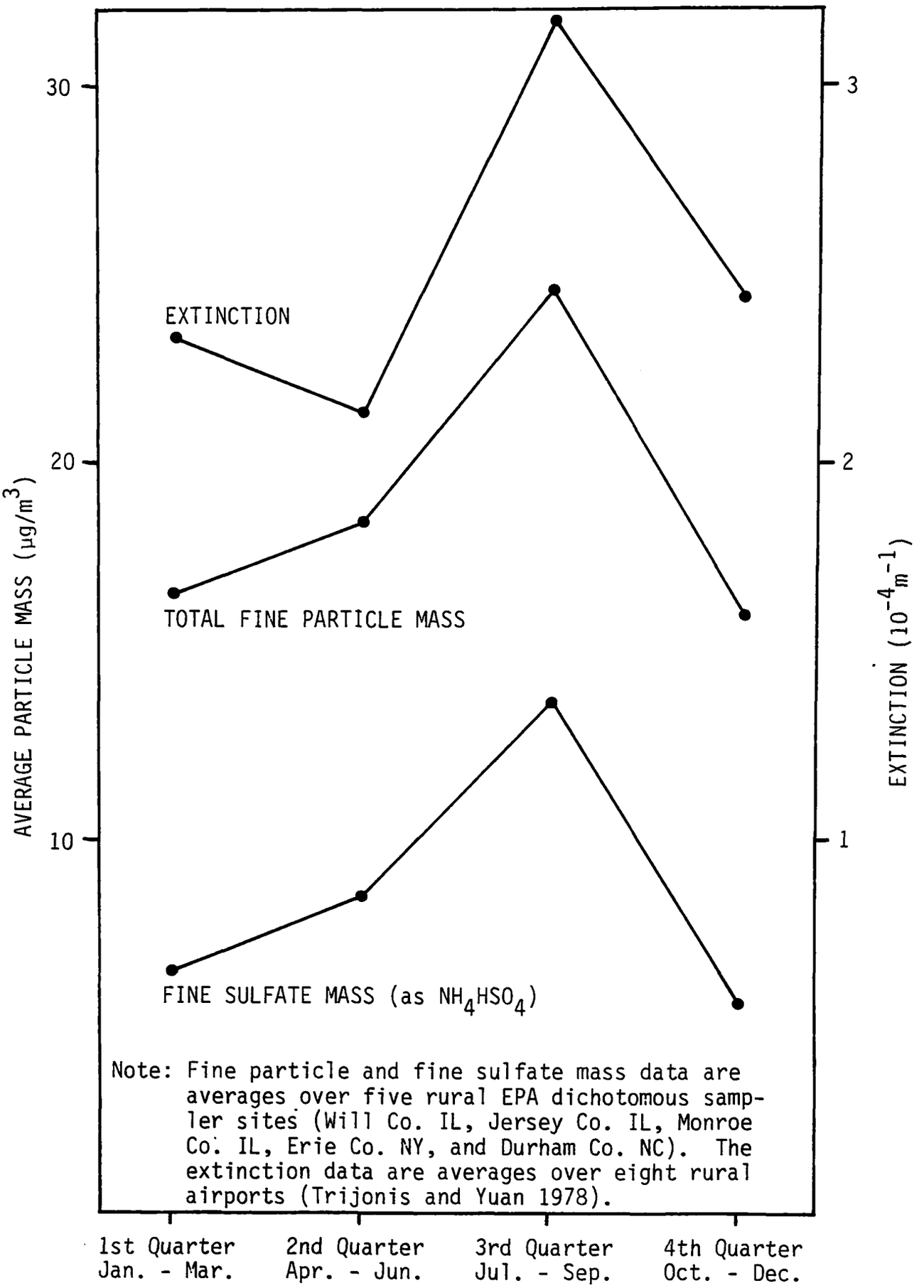

Figure 3. The seasonal patterns in sulfates, fine particle mass, and extinction for rural areas of the East.

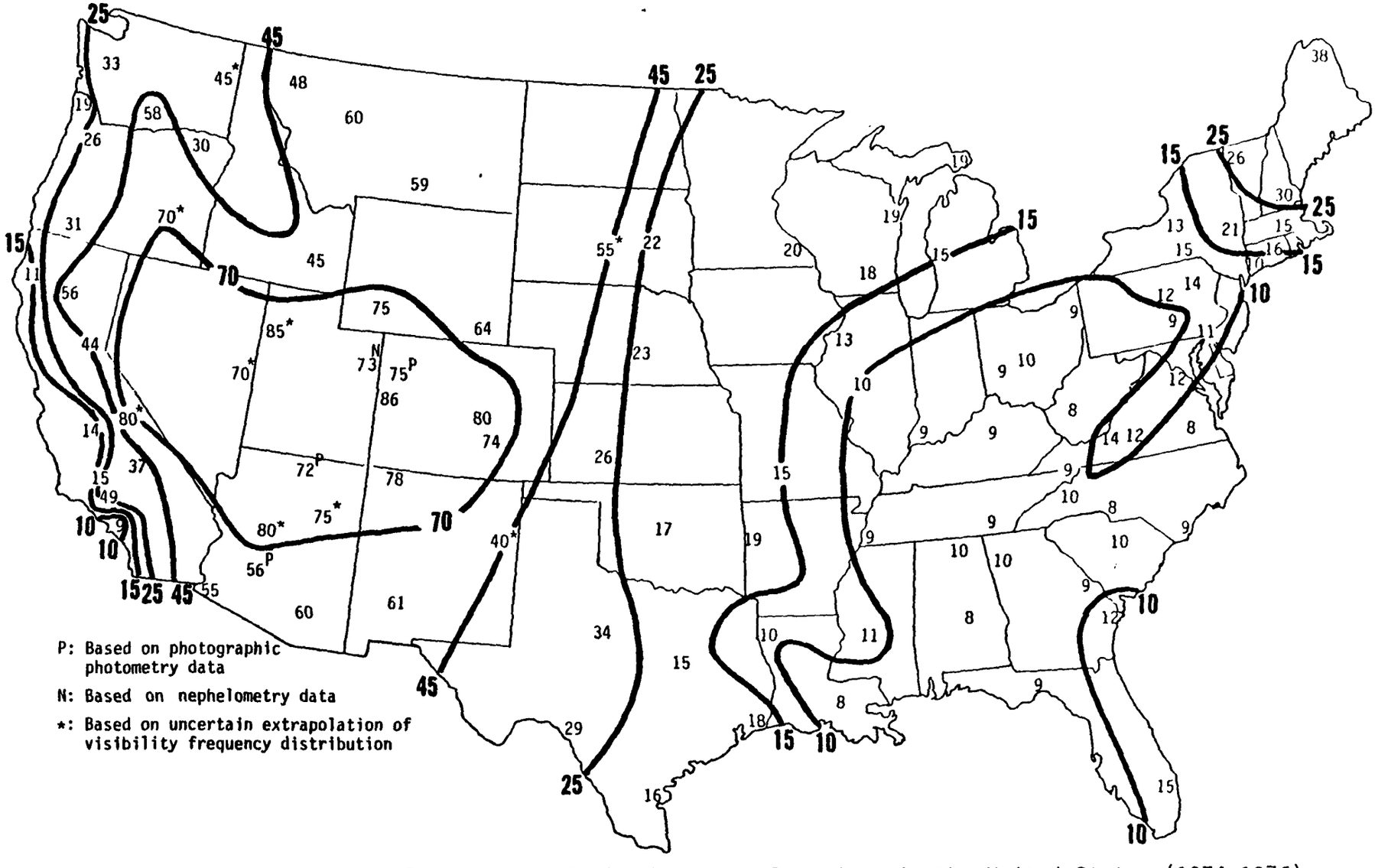

Figure 4. Median summer visual range at suburban/nonurban locations in the United States (1974-1976).

conversations with thirty aerosol researchers to solicit data, comments, and suggestions regarding our calculations. These researchers are listed in the acknowledgement section of this paper.

Table 3 presents the end result of our investigation -- a model ambient aerosol for Eastern natural background conditions. The error bounds in Table 3 are much larger (in a relative sense) than the error bounds in Table 2, because estimating natural background conditions is a very precarious undertaking for many of the aerosol components. As a reflection of the uncertainties, we have listed the values in Table 3 as fractions (e.g. ¼ or ½ $\mu g/m^3$) rather than as decimals. Our specific reasonings and calculations for the various aerosol components are described in the paragraphs that follow.

Two independent approaches suggest that natural background sulfate concentrations in the East are very low. One approach concerns ambient measurements made in the Southern Hemisphere which has an order of magnitude less man-made SO_x emissions than the Northern Hemisphere (Cullis and Hirschler 1980). Most of these measurements have been made in areas that, like the Eastern U.S., are humid and densely vegetated. Lawson, Winchester, and coworkers have collected data on fine sulfate concentrations at numerous locations in the Southern Hemisphere and have recently published an overview (Lawson and Winchester 1979). Table 4, a summary of that overview, suggests that natural background fine sulfate concentrations are on the order of 0.1 to 0.5 $\mu g/m^3$. Even these very low values could represent overestimates because anthropogenic influences at remote subtropical Southern Hemisphere locations may be non-negligible (Lawson 1981).

TABLE 3. MODEL AMBIENT FINE AEROSOL, ANNUAL MEAN FOR NATURAL BACKGROUND CONDITIONS IN THE EAST.

COMPONENT	MASS CONTRIBUTION* (g/m^3)
Sulfate (as NH_4HSO_4)	½ ± ½
Organics	2 ± 2
Elemental Carbon	¼ ± ¼
Crustal Material	½ ± ½
Nitrates (as NH_4NO_3)	¼ ± ½
Non-water Other	½ ± ½
Water	1½ ± 1
TOTAL	5½ ± 2½**

*Mass of particles less than 2.5 microns in size under ambient conditions (RH ∿ 70-75%).

**Total error is calculated as the root-mean-square of the individual errors.

NATURAL BACKGROUND VISUAL RANGE (See Text): 60 ± 30 miles.

TABLE 4. SUMMARY OF AVERAGE FINE SULFATE CONCENTRATIONS AT SOUTHERN HEMISPHERE LOCATIONS (Lawson and Winchester 1979).

LOCATION	AVERAGE FINE SULFATE CONCENTRATION EXPRESSED AS NH_4HSO_4 (in $\mu g/m^3$)
Five remote locations in South America	0.04 to 0.16
Four remote locations in South America possibly under the direct influence of sea spray or vegetative burning	0.20 to 0.50
One nonurban location in West Africa	0.6
One nonurban location in Samoa	0.2

The second approach involves emissions estimates for sulfur oxides. Several recent publications have concluded that man-made SO_x emissions dominate natural SO_x emissions in the Eastern United States (Adams et al. 1980; Galloway and Whelpdale 1980; Henry and Hidy 1980; Rice et al. 1981). Based on measured emission rates, Adams et al. calculated that terrestrial biogenic sources contribute only 1 to 2% of the sulfur burden in the Eastern U.S. Considering both terrestrial and marine influences, Galloway and Whelpdale calculated a 4% natural sulfur contribution for the Eastern U.S. Using our model ambient aerosol for rural Eastern areas in Table 3 (including 9 $\mu g/m^3$ of sulfate), a 4% natural contribution represents less than 0.4 $\mu g/m^3$ of natural background sulfate. This value agrees closely with the Lawson and Winchester data reported in the previous paragraph.

As our final estimate of natural background sulfate aerosol concentration in the East, we have chosen $\frac{1}{2}$ $\mu g/m^3$. The error bound is $\pm \frac{1}{2}$ $\mu g/m^3$.

Estimating natural background levels for fine organic aerosols in continental areas is a very uncertain procedure. Some of the uncertainties are highlighted in recent reviews by Duce (1978) and Hahn (1980). One major problem is the lack of data for truly remote continental areas (i.e. in the Southern Hemisphere). There are several data sets for remote marine areas (Hoffman and Duce 1974, 1977; Ketseridis et al. 1976; Barger and Ganett 1976; Eichmann et al. 1979; Chesselet et al. 1981). Using adjustment factors developed by Duce (1978) and Hahn (1981), (total organic aerosol $\approx$ twice ether extractable organic aerosol, total organic aerosol $\approx$ 1.5 times organic carbon, and fine organic aerosol $\approx$ 0.8 times total organic aerosol), these various data sets yield a very consistent value of 1 $\mu g/m^3$

for fine organic aerosol concentrations in remote marine areas. This value, however, may seriously underestimate natural continental organic concentrations, because the organic aerosols from major natural continental sources (e.g. primary plant wax aerosols, secondary terpenic aerosols, etc.) might not be adequately represented in remote marine areas. On the other hand, there could be a slight compensating overestimate due to the presence of transported anthropogenic organic aerosols into remote marine areas. Because of these confounding and unknown effects, it does not appear reasonable to infer natural continental organic aerosols from the measurements made in remote marine areas.

Our estimate of natural fine organic aerosol concentrations in the East, 2 $\mu g/m^3$, is based on two considerations. First, Duce (1978) and Hahn (1980) interpret available information as suggesting a remote continental background of 1½ to 2¼ $\mu g/m^3$ of organics. Second, we calculate a value of 2 $\mu g/m^3$ with a crude "tracer" calculation based on the following assumptions: (1) there are 4 $\mu g/m^3$ of total organic aerosol and 1 $\mu g/m^3$ of elemental carbon aerosol in rural areas (see Table 2); (2) the ratio of organic aerosol to elemental carbon aerosol is 2:1 in urban Eastern areas (Wolff et al. 1980); and (3) elemental carbon (essentially all man-made) can be used as a tracer for the contribution of man-made organics according to the 2:1 ratio. The error bounds for our estimate ($\pm$ 2 $\mu g/m^3$) are large because emission rate calculations suggest potentially much greater concentrations of plant waxes and terpene derivatives (Duce 1978; Went 1960; Rasmussen and Went 1965; Beauford et al. 1977), whereas certain ambient studies find little or no direct evidence of significant contributions from natural organic aerosols (Crittenden 1976; Daisey et al. 1979).

Elemental carbon (soot) constitutes only 1 $\mu g/m^3$ of our model rural ambient aerosol in Table 2. Two approaches suggest that this component is basically anthropogenic. The first approach involves emission data for elemental carbon. The only significant natural source of elemental carbon in the East is wildfires. Comparing wildfire soot emissions to total soot emissions from prescribed burning, diesels, gasoline vehicles, aircraft, solid waste incineration, and gas/fuel-oil/coal/wood combustion (EPA 1973, 1976, 1979; Muhlbaier and Williams 1981; Muhlbaier 1981; Cass et al. 1981), we estimate that wildfires contribute only a small fraction of total elemental carbon emissions. The second approach is based on a "lead tracer" calculation. Both lead and soot in urban areas are essentially all man-made. Lead concentrations in rural Eastern areas are also generally assumed to be man-made. That lead and soot both exhibit approximately the same dilution factor from urban to nonurban areas of the East -- a factor of about 5-10 (EPA 1972, 1981; Stevens et al. 1980; Wolff et al. 1980) -- suggests that soot concentrations in nonurban areas are also mostly man-made. For our natural background aerosol in Table 4, we have assumed an elemental carbon concentration of $\frac{1}{4} \pm \frac{1}{4}$ $\mu g/m^3$. This may be an overestimate, but the errors are not important because elemental carbon is a very small component of the aerosol.

Crustal material accounts for 1 $\mu g/m^3$ of the model fine rural aerosol in Table 2. This component is composed of fly ash and/or the lower tail of the soil dust size distribution. Based on a study of the spatial patterns, meteorological dependencies, and size distributions of the crustal elements at nonurban sites near St. Louis (Trijonis et al. 1980), we conclude that this component is predominantly soil dust rather than fly ash. It is

difficult, if not impossible, to calculate how much of the fine soil dust is natural wind blown dust versus anthropogenic dust (e.g. dust raised by traffic, construction, and agriculture or by wind action over surfaces disturbed by man). Arbitrarily assuming that half is related to man-made activities, our estimate for the natural background concentration of fine crustal particles in the East is $\frac{1}{2} \pm \frac{1}{2}$ $\mu g/m^3$. The error in this assumption is not critical because fine crustal material is a small component of the aerosol.

Nitrate aerosols also contribute 1 $\mu g/m^3$ to the model fine Eastern aerosol in Table 2. Recent measurements indicate that fine nitrate concentrations in remote continental and marine areas of the Northern Hemisphere average approximately .05 to .2 $\mu g/m^3$ (Huebert and Lazrus 1980). Adopting a somewhat conservative approach, we will assume that average natural background nitrate concentrations in the East are $\frac{1}{4} \pm \frac{1}{4}$ $\mu g/m^3$. Again, the errors in this assumption are not important because nitrate is a small component of the fine aerosol.

Natural background concentrations of the "other non-water" component should be low. In fact, it is hard to imagine significant contributors to the average fine natural aerosol that are not sulfates, organics, soot, crustal material, nitrates, or water. The lower tail of the sea-salt size distribution would be significant in coastal areas but not of great importance averaged over the East. We will arbitrarily assume that the average natural background level for the "other non-water" category is $\frac{1}{2} \pm \frac{1}{2}$ $\mu g/m^3$.

Table 2 indicates that the rural Eastern fine aerosol currently contains 11 $\pm$ 5 $\mu g/m^3$ of water. To estimate the amount of water attached to the natural background fine aerosol, we consider two alternative hypotheses.

First, we assume that all of the water is attached to hygroscopic sulfate and nitrate aerosols, and that the amount of water in the natural aerosol should be in proportion to the sulfate and nitrate remaining. This hypothesis yields an estimate of 1 $\mu g/m^3$ for "natural" water (with a range of 0 to 2 $\mu g/m^3$ considering the error bounds in Tables 2 and 3). As an alternative, we assume that the water is attached equally to all components of the non-water aerosol, and that the amount of water in the natural aerosol should be in proportion to the total fine aerosol mass remaining. This second hypothesis yields an answer of 2½ $\mu g/m^3$ (with a range of 0 to 5 $\mu g/m^3$ considering the error bounds in Tables 2 and 3). The truth should be somewhere between our two alternative hypotheses. The first hypothesis underestimates natural aerosol water because at least some small fraction of the water should be attached to non-sulfate, non-nitrate particles, and because the natural sulfate aerosol may be more acidic (and therefore more hygroscopic) than existing sulfate aerosols (Mueller 1981). The second hypothesis is not reasonable because much more water should be attached to hygroscopic aerosol components than to hydrophobic aerosol components. We will assume that the natural fine aerosol contains $1\frac{1}{2} \pm 1$ $\mu g/m^3$ of water.

As an aside, we should comment on a question that we have sometimes heard raised: "If man-made sulfates were eliminated, wouldn't some of the water associated with the man-made sulfates have a tendency to become attached to the remainder of the aerosol?" The answer to this question is "No" because transferring water from the aerosol to the gas phase produces essentially no change in relative humidity. Simple calculations demonstrate that water in the gas phase is typically orders of magnitude greater than water in the particulate phase.

Table 3 indicates that the model fine aerosol for average natural background conditions in the East adds up to 5½ ± 2½ $\mu g/m^3$. The two largest components are organics (2 ± 2 $\mu g/m^3$) and water (1½ ± 1 $\mu g/m^3$). Each of the other individual components is estimated to contribute ½ $\mu g/m^3$ or less to the total.

The fine aerosol concentrations in Table 3 can be used to estimate natural background visibility levels in the East. Noting that median visual range averaged over the East is currently about 13 miles (see the first section of this paper as well as Allard and Tombach 1980), assuming that dry sulfate has a mass scattering efficiency 1.5 times greater than other dry aerosol components and that water has a mass scattering efficiency 1.7 times greater than dry sulfate (see earlier discussion and references), taking into account absorption by elemental carbon (Ferman et al. 1981; Groblicki et al. 1981), and taking into account Rayleigh (blue-sky) scatter by air molecules, we calculate that median visual range would be 60 ± 30 miles under average natural background conditions in the East.

We conceived a crude method of checking this estimate of natural background Eastern visibility based on an inter-site regression analysis. Specifically, we regressed annual median visibility levels at various rural Eastern sites (from Figure 2) against annual mean sulfate levels (from NASN data) and annual mean relative humidity (from NOAA 1977). Following the physical principles discussed by Cass (1979) and Trijonis and Yuan (1978), the regression equations used extinction coefficient (3.9 ÷ visual range) as the dependent variable and "sulfate/(1-RH)" as the independent variable. By plugging estimated natural background sulfates (½ $\mu g/m^3$) and average

Eastern relative humidity (70%) into this regression equation, we might obtain a crude approximation of natural background visual range. This approach should underestimate natural background visual range because non-sulfate anthropogenic aerosols are not explicitly discounted for; however, this difficulty should be mitigated by the fact that anthropogenic sulfates are colinear (correlated) with other anthropogenic aerosols.

The spatial regression method yielded a natural visibility estimate of 30-40 miles, not bad agreement with our previous value of 60 ± 30 miles. There were indications, however, that the spatial regression analysis was too crude to be accepted as reliable. There was a great amount of scatter in the data points, the regression coefficients were unstable, and the results depended significantly on which sites were included (e.g. on whether only nonurban or both nonurban and suburban sites were included, or on which geographical areas were included).

Although the spatial regression analysis was not a reliable method of checking our estimate of natural background visibility, it did provide insights regarding an important concept -- the geographical distribution of natural visibility levels in the East. Our estimates of natural background levels for fine particles and visibility are intended to be spatial averages for the entire East. It is important to recognize, however, that natural background levels of fine particles and visibility might exhibit substantial spatial variations over the East. For example, one might expect organic aerosol concentrations to be greater in the south than in the north. This possibility is, in fact, strongly suggested by our inter-site regression analysis which indicates lower natural background visibility for the south

than for the north if the data are divided geographically. Also, one would expect natural background visibility to be lower for coastal areas than for inland areas because of higher relative humidity (NOAA 1977) and greater sea salt concentrations in the coastal areas. Insufficient data are available to quantify the geographical variations of natural background fine particle/visibility levels in the East; it is nevertheless important to acknowledge that spatial variations would exist under natural conditions.

LONG-TERM VISIBILITY TRENDS

As part of this investigation, we also considered the possibility of checking our estimate of natural background Eastern visibility (60 ± 30 miles) by comparing long-term visibility trends with historical trends in anthropogenic emissions. This comparison might suggest a relationship between visibility and emissions that could be extrapolated back to zero man-made emissions. Sulfur oxides would be the most appropriate emission variable for the comparison because sulfates and associated water constitute the predominant visibility reducing component of the Eastern aerosol (see discussion and references in the previous section, "Existing Fine Particle Concentrations"). Gschwandtner et al. (1981) have recently compiled sulfur oxide emission trends for the Eastern U.S. for the period 1950 to 1978. Also of relevance, estimates of national coal consumption are available from the early 1800s to 1970 (Bureau of Census 1975; Husar and Holloway 1980)

For the long-term visibility trend data, we selected eight suburban/rural Eastern airports that have adequate visibility marker systems, have

data back to the early 1930s, and have not undergone major relocations. These airports are Evansville IN, Louisville KY, Albany NY, Charlotte NC, Dayton OH, Allentown PA, Nashville TN, and Burlington VT (see Figure 5). Actually, in a survey of all suburban/rural airports in the East, we found that few, if any, airports besides these eight met all our criteria regarding marker systems, long-term data availability, and site relocations. Even these eight airports are not ideal because nearly all of them have undergone at least some (minor) relocations.

In the long-term trend analysis, we compiled airport visibility data for three-year periods corresponding to the second to fourth year of each decade (e.g. 1932-1934, 1942-1944, etc.). We did not consider every year of each decade because we lacked sufficient resources for the effort of compiling hard-copy visibility data (for the 1930s and 1940s, we had to manually transcribe hard-copy records from the National Climatic Center archives in Asheville NC). The second to fourth year of each decade seemed to be a logical choice for two reasons: (1) the visibility records at most airports started around 1932, and (2) the historical cycle in coal usage exhibits maxima and minima during these years (e.g. a Great Depression minimum in 1932-1934, a war-time maximum in 1942-1944, and clean-fuel switch minima in the early 1950s and early 1960s) (Husar and Holloway 1980).

From the early 1940s to the early 1970s, the visibility trends at seven of the airports agree qualitatively with one another and with historical SO_x emission trends. As shown in Figure 6, visibility increased at these seven airports from the 1940s to the 1950s in agreement with the substantial post-war decrease of national coal consumption (Husar and Holloway 1980). Visibility at the seven airports decreased from the 1950s to the

Figure 5. Airports used to analyze long-term visibility trends.

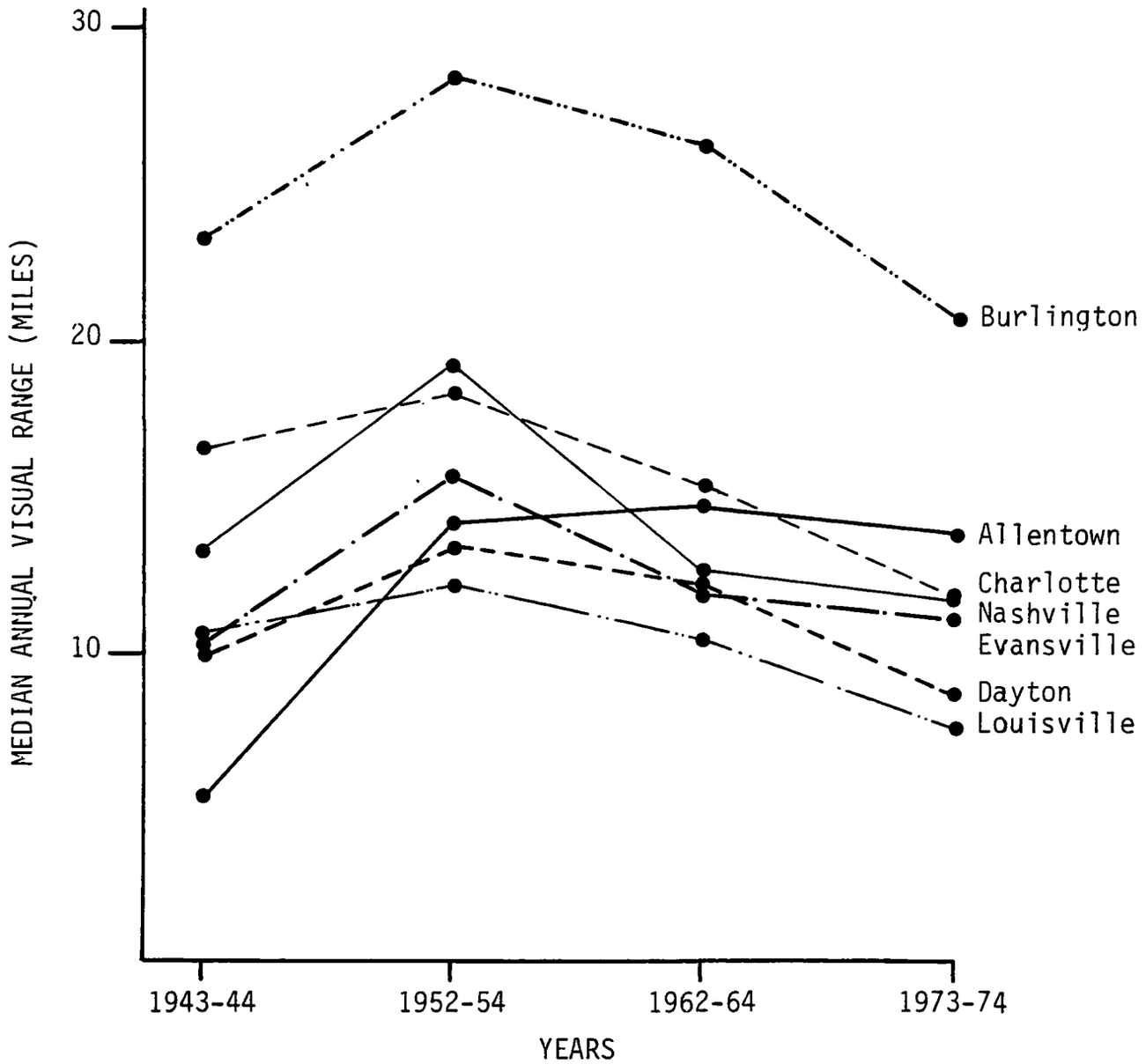

Figure 6. Visibility trends at seven Eastern airports from the early 1940s to the early 1970s.

1970s in agreement with the increases in estimated Eastern SO_x emissions (Gschwandtner et al. 1981) and national coal consumption (Husar and Holloway 1980) during that period.

We caution the reader, however, that the above agreement should be regarded with some skepticism. For one, the available emissions trend data are questionable. For example, although the Eastern SO_x emission estimates (Gschwandtner et al. 1981) and national coal consumption estimates (Husar and Holloway 1980) both show some increase from the early 1950s to the early 1970s, the former indicates a much larger increase than the latter. Part of this discrepancy is probably due to inaccuracies in the Eastern SO_x emission estimates; Gschwandtner (1981) has stated that the data he used to compile emission trends are less than adequate prior to 1960, and that certain corrections must be made to his 1950s values. Part of the discrepancy may also reflect the facts that the coal consumption data do not necessarily track SO_x emissions (due to changes in sulfur content, processes, etc.), and that the coal consumption data represent national rather than Eastern figures. In any case, because of this discrepancy, we also feel less confident in using the post-war decrease of national coal consumption as a measure of post-war decreases in Eastern SO_x emissions.

As a second caution, we note that meteorology may have played a significant role in determining visibility trends. Although visibility trends stratified by meteorological parameters tend to be similar to the overall visibility trends presented in Figure 6 (Trijonis and Yuan 1978), we cannot rule out the possibility that the qualitative agreement between Figure 6 and historical SO_x emission trends is a fortuitous one produced by meteorological

fluctuations. The potentially confounding effect of meteorology seems all the more important when one considers that SO_x emissions over the past five decades have fluctuated only about ± 30% compared to the 50-year average (Husar and Holloway 1980).

The third caution concerns the tenuous nature of historical trend studies based on airport visibility data. It has been found that airport data are of generally good quality for studying geographical/seasonal/meteorological patterns in visibility but are of questionable quality with respect to historical trend analysis (Trijonis 1981). The major problem is that, in historical trend analysis, one is usually dealing with long-term visibility changes on the order of 30% or less. Changes in median visibility of 10 to 20% can also be produced artificially by modifications in visibility reporting practices. One way of circumventing this problem is to examine data from numerous airports and to verify that the trends are consistent among the airports. In this regard, we can be somewhat encouraged that seven of our eight airports are in qualitative agreement.

In addition to the general uncertainties in airport visibility data discussed in the previous paragraph, we must note three specific problems in our particular airport data sets. First, the visibility trends at Albany NY do not agree with the trends at the other seven locations. In fact, Albany shows exactly the opposite pattern -- decreasing visibility from the 1940s to the 1950s and increasing visibility from the 1950s to the 1970s. Increasing visibility from the 1950s to the 1970s in New York State has been previously noted by Husar et al. (1979); they attributed this atypical pattern to the fact that New York, unlike most other Eastern areas, underwent decreases in coal usage and SO_x emissions from the 1950s to the 1970s.

Despite an apparent explanation for the atypical visibility trends at Albany, the contradiction represented by the Albany trends remains an important caveat to our analysis. This caveat is strengthened even further by the results of Sloane (1980), who found that some rural sites in the Appalachian area did not show the visibility decreases exhibited by most other rural Eastern sites from 1948 to 1978. Second, we believe that the large increase in visibility (decrease in extinction) at Allentown from the 1940s to the 1950s is more of a data quality artifact than a real effect associated with decreased post-war emissions. The visibility reporting system at Allentown during the 1940s was extremely unusual and therefore highly questionable. Third, the visibility trends at the various airports from the 1930s to the 1940s are a hodgepodge. Half of the airports show improving visibility (of widely varying degrees) from the 1930s to the 1940s, while half show deteriorating visibility (of widely varying degrees). We think that this reflects problems with the quality of the 1930s airport visibility data. Personnel at the National Climatic Center stressed to us that visibility reporting practices were at an infancy stage during the early 1930s and that good consistency was not attained until the 1940s. In fact, we found some reporting codes (involving both letters and numbers) for visual range during the 1930s that none of our contacts at NCC could interpret. Although the lack of consistent visibility trends from the 1930s to the 1940s probably reflects data quality problems, it could also be interpreted as a contradiction to the correspondence between historical visibility trends and SO_x emission trends. Specifically, national coal consumption data suggest that the 1932-1934 period had significantly less SO_x emissions than the 1942-1944

period; on the other hand, the available (albeit poor) airport data for that period show no clear deteriorating trend in visibility from the 1930s to the 1940s.

Because of the uncertainties in the visibility and emission data, it is not presently worthwhile to attempt a quantitative comparison of visibility trends and SO_x emission trends for the purpose of estimating natural background visibility. Such a quantitative analysis may become feasible in the future as some of the uncertainties are resolved. The most immediate problem to be solved is the accurate estimation of SO_x emission trends prior to 1960. Even if this problem is solved, however, a good quantitative analysis may be precluded by the uncertainties in visibility trend data and by the fact that SO_x emissions have changed only moderately (± 30%) over the past five decades.

SEASONAL PATTERN OF HISTORICAL VISIBILITY TRENDS

In a previous section, we showed that sulfates and total fine particle concentrations currently exhibit pronounced maxima and that visibility currently exhibits a pronounced minimum during the summer season (3rd calendar quarter) in the East. It is of interest to examine this seasonal pattern historically. Fortunately, the data quality problems associated with historical trend analysis of airport visibility data are essentially eliminated when one considers relative visibility levels among seasons. Artificial trends produced by site relocations and by changes in reporting practices should affect all seasons equally and should cancel out if one just compares relative visual range among seasons.

Figure 7 illustrates historical trends in the ratio of median third quarter visibility to median visibility for the other three calendar quarters. Trends are presented for all eight study sites from the early 1930s to the early 1970s. Figure 7 shows that, during the 1930s, 1940s, and 1950s, summertime visibility exceeded visibility for the remainder of the year by a factor of 1.0 to 1.5. From the early 1950s to the early 1970s, however, summertime visibility declined precipitously relative to visibility for the remainder of the year so that, by the early 1970s, summer had become a season of distinctly poor visibility.

Many other researchers have noted a strong decline in summertime visibility (Miller et al. 1972; Trijonis and Yuan 1978; NRC 1979; Husar et al. 1979; Sloane 1980) and a corresponding increase in summertime sulfate concentrations (Trijonis 1975; EPA 1975; Altshuller 1976; Frank and Possiel 1976; NRC 1979) in rural Eastern areas from the 1950s to the 1970s. This phenomenon has been attributed to the rapid growth of summertime SO_x emissions from coal-fired power plants due to air conditioning demands (Holland et al. 1977; Husar et al. 1979; Sloane 1980). Also, summertime visibility should be particularly sensitive to SO_x emissions because photochemical processes are important in the conversion of sulfur dioxide emissions to sulfate aerosols, and because solar radiation is most intense in the summer.

SUMMARY AND CONCLUSIONS

Existing visibility in rural areas of the Eastern U.S. is rather low. For example, in contrast to the 70-85 mile median visual range in the

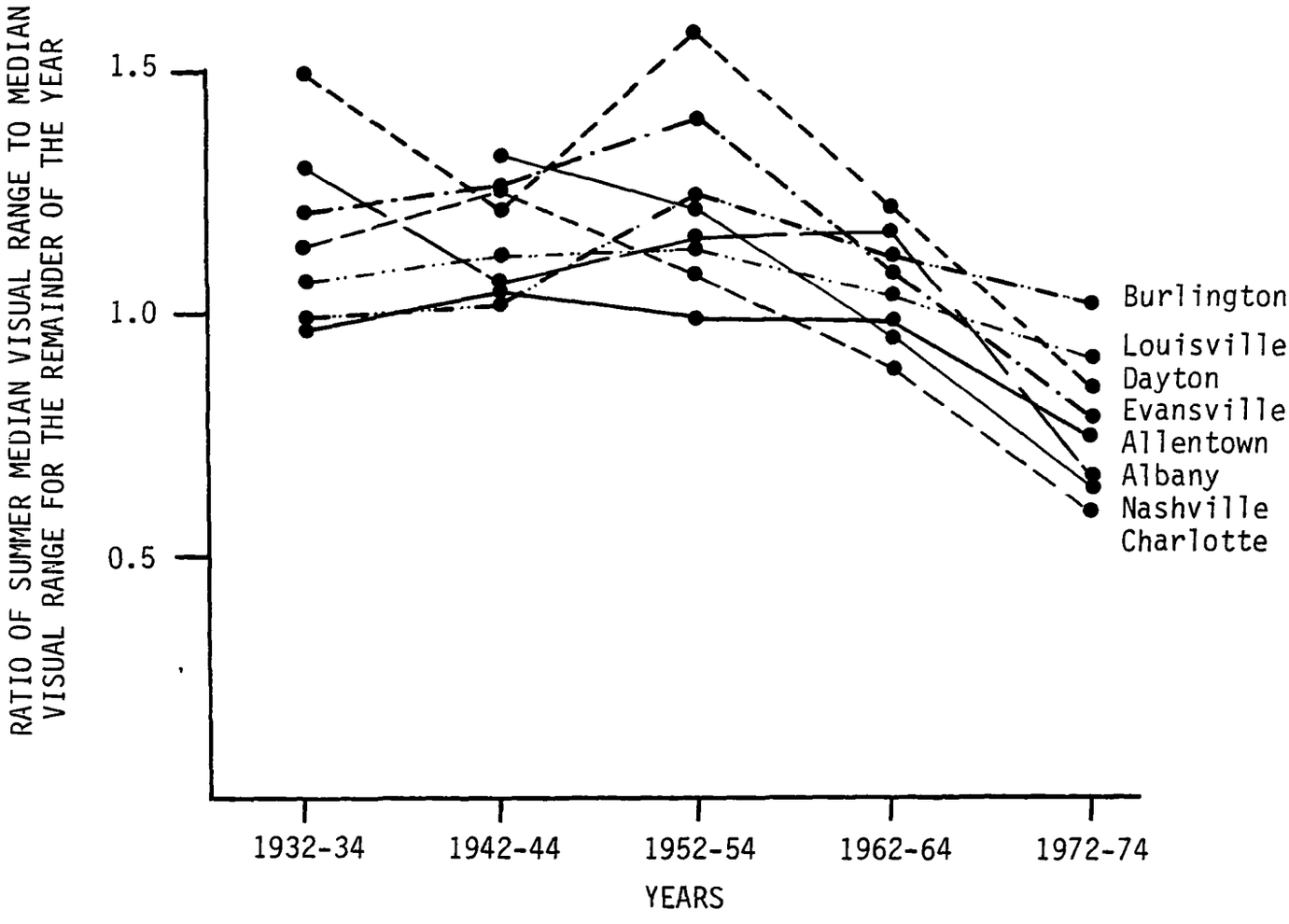

Figure 7. Historical trends in the ratio of summertime visibility to visibility during the remainder of the year.

mountainous Southwest, annual median visual range is only 10-15 miles over most of the area south of the Great Lakes and east of the Mississippi. Some parts of the East, however, such as New England and the eastern slope of the Appalachians, experience more moderate visibility levels (on the order of 15-35 miles).

Visibility in the East is essentially controlled by ambient fine particle concentrations. Based on data from eight monitoring programs and on information concerning water content of aerosols, we conclude that -- annually and spatially averaged -- there is about 29 $\mu g/m^3$ of ambient fine aerosol in rural Eastern areas. The components are water (11 $\mu g/m^3$), sulfates (9 $\mu g/m^3$), organics (4 $\mu g/m^3$), soot (1 $\mu g/m^3$), crustal material (1 $\mu g/m^3$), nitrates (1 $\mu g/m^3$), and other (2 $\mu g/m^3$). Because most of the water is probably attached to hygroscopic sulfate aerosols, sulfates and associated water might be interpreted as a single predominant component of the fine aerosol. This would explain the high statistical correlations that have been observed between sulfate concentrations and visibility reduction.

Fine particle concentrations and visibility in the East currently exhibit strong seasonal patterns. Specifically, sulfates and total fine particles reach a pronounced maximum in the summer (third) quarter, when visibility reaches a pronounced minimum. This seasonal pattern is apparently a rather recent phenomenon historically; data from the 1950s and prior decades indicate that summer used to have significantly higher visibility than the remainder of the year.

Under natural background conditions, we estimate that rural Eastern areas would have an average fine aerosol concentration of $5\frac{1}{2} \pm 2\frac{1}{2}$ $\mu g/m^3$. The

largest components would be organics ($2 \pm 2\ \mu g/m^3$) and water ($1\frac{1}{2} \pm 1\ \mu g/m^3$). Sulfates would contribute only about $\frac{1}{2}\ \mu g/m^3$ under natural conditions. This natural ambient fine aerosol would produce an average visual range of 60 ± 30 miles.

It is not currently possible to derive natural background visual range from long-term visibility trend data because of limitations in historical emission estimates, uncertainties in airport visibility trend data, and the confounding influences of meteorology. Even if some of these uncertainties are resolved, quantitative analyses of long-term visibility trends will be hindered by the fact that SO_x emissions have varied only ± 30% over the past five decades. The most promising method of checking our estimates of natural background conditions would be to conduct field studies in remote continental areas of the Southern Hemisphere. Such field studies should include simultaneous measurements of fine particle mass, fine particle chemical composition, and atmospheric optical parameters (visibility).

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10. PROGRAM ELEMENT NO.

11. CONTRACT/GRANT NO.

12. SPONSORING AGENCY NAME AND ADDRESS

Monitoring and Data Analysis Division
Office of Air Quality Planning and Standards
US Environmental Protection Agency
Research Triangle Park, NC 27711

13. TYPE OF REPORT AND PERIOD COVERED

14. SPONSORING AGENCY CODE

15. SUPPLEMENTARY NOTES

EPA Project Officers: E. L. Martinez and J. Bachmann

16. ABSTRACT

Existing and natural background levels of visibility and fine particles are investigated for nonurban areas of the Eastern U.S. Analysis of data for 100 airports nationwide indicates that nonurban areas of the East experience relatively low visibilities. Eastern rural areas generally show annual median visual ranges of 10-15 miles. Data from eight monitoring programs indicate that ambient fine (<2.5 μm) particle concentrations presently average approximately 29 $\mu g/m^3$ in the rural East. Main components are water, sulfates and organics. Currently seasonal sulfate and fine particle levels peak in summer, when visibility is lowest. Prior to the 1960's, visibility was distinctly higher in summer than the rest of the year. An investigation of natural background conditions suggests that natural fine particle concentrations would average $5\frac{1}{2} \pm 2\frac{1}{2}$ $\mu g/m^3$ in the East, mostly composed of organics and water. Natural background visual range for the East is estimated to be 60 ± 30 miles.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS *(This Report)*	21. NO. OF PAGES
		46
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