INADVERTENT MODIFICATION OF VISIBILITY IN THE VICINITY OF A MAJOR METROPOLITAN AREA

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INTRODUCTION

Urban areas have always been suspect in the production of large amounts of aerosols and accompanying visibility restrictions; recently this has been reaffirmed in the case studies by several investigators (Shea and Auer, 1976; Lyons and Rubin, 1976; White et al., 1976). This note briefly reports on some additional information on the evolution of visibility anomalies and the accompanying aerosol size spectra derived from cloud condensation nucleus (CCN) measurements in the downwind plume of St. Louis.

PROCEDURES

The monitoring of real time output from the airborne meteorological system (Endsley and Knowlton, 1973) aboard the University of Wyoming's Queen Air Research Aircraft was used to insure a constant traverse within the urban plume. Transport winds, derived from a Doppler navigation system, and Aitken nucleus concentrations were the principal parameters monitored, although other meteorological parameters along the flight experience were also used. Once the mean transport winds within the mixing layer were predicted by antecedent winds aloft observations and confirmed by the aircraft, the aircraft would fly downstream for an appropriate distance corresponding to approximately 5 hours of travel time for a parcel traveling with the mean transport wind. Then the aircraft would reverse heading and fly toward the urban area. Visibilities were monitored along the heading each way and CCN samples were collected sequentially in metalized mylar bags on the inbound leg.

These air samples were processed in a thermal diffusion chamber according to generally accepted procedures and precautions (Fitzgerald, 1970; Saxena et al., 1970; Fitzgerald, 1972; Squires, 1972). Corrections were made for the diffusion of nuclei to the walls of the bags. Concentrations of CCN at activating supersaturations from 0.25 to 1.5% were derived and expressed in the form

$$n = c\left(\frac{S}{S_0}\right)^k \qquad (1)$$

where n is the concentration (cm^{-3}) of CCN active at the supersaturation, S, S_o, is arbitrarily set at 1%, and the constants c and k determine the properties of the spectra.

Light scattering aerosol size spectra were derived for various times downwind utilizing the following procedures and components. Empirical determination of activating supersaturations for NaCl particles (Ruskin and Kocmond, 1971; Katz and Kocmond, 1973) and natural aerosols (Ruskin and Kocmond, 1971) between 0.05 and 0.2μm diameter has been made. Recent studies (Hindman et al., 1975) of aerosols within and outside of a paper mill plume confirm the previous empirical expressions. A representative form of this expression is:

$$D_p = 7.5 \times 10^{-2} \left(\frac{S}{S_o}\right)^{-0.6} \quad (2)$$

where D_p is the diameter in microns.

Now if the concentration, N, of aerosols between 0.05 and 0.2μm within and outside our urban plume act as CCN, according to (1), then n = N and (1) and (2) can be combined to yield an expression of the aerosol size distribution

$$N = A\left(\frac{D_p}{D_o}\right)^b \quad (3)$$

where N is the concentration (cm^{-3}) of aerosols with diameter equal to or greater than Dp, D_o is arbitrarily set at 1μm and A and b are descriptive constants of the spectra.

Aitken nucleus concentrations were determined with the Rich 100 continuous Aitken nucleus counter manufactured by Environment One. Instrument description, principles of operation, and test comparison have been provided (Saxena et al., 1973). The total Aitken nuclei number was used to extend the log diameter-number distribution (e.g., Figure 1) to the smallest diameters under the assumption that the diameter interval of the Aitken nuclei is Δlog D = 1.

A PMS[1] axially scattering spectrometer probe (ASSP) was used to continuously measure in situ the number of aerosols between 1.5-2.5μm by measuring the amount of light energy they scatter out of a laser beam.

Thus, aerosol size distributions can be generated from the Aitken nucleus concentrations, derived size spectra of the CCN and the ASSP data.

The term "visibility" is used in this paper in its meteorological sense, which is more properly "visual range" or the greatest distance

[1]Particle Measuring Systems, Incorporated, Boulder, Colorado.

at which prominent objects can be seen. Estimates of visibility were made from the aircraft flying in either direction parallel to the orientation of the urban plume. Slant range and horizontal visibilities have been equated here since all visual estimates were performed at altitudes at or below 600 m (2000 ft) AGL. Visibility measurements were made between 1100-1500L, preferably with the sun not behind the observer but in the field of vision. Objects for visibility marks were as dark as possible, silhouetted against the horizon sky. Glittering objects were avoided. Dull-colored objects coming into the field of vision often served as a measure of visibility.

Experiments were conducted in the mid-day mixing layer (relative humidity averaging 65%) characterizing fair weather conditions during July-August 1974-75. Visibility and aerosol data presented represent values believed typical for the urban plume conditions and are not knowingly biased toward a particular industrial site. The transport time was computed by dividing the downwind distance along the mean wind direction by the wind speed in the mixing layer. The wind speed exceeded 6 m sec^{-1} in all cases. All transport times are with respect to the Gateway Arch, centroid of the urban-industrial activity in St. Louis.

RESULTS

The results discussed present a synthesis of analyses of five case studies with simultaneous aerosol and visibility data, plus one case study with only aerosol data.

Figure 1 shows the changes in visual range synthesized from 6 case studies as a function of the downwind transport time. Also displayed in Figure 1 are time sections of four characteristics of the downwind aerosol spectra: ratio of downwind to upwind CCN concentrations, active at 0.5% supersaturation; Aitken nucleus concentrations of particles between 1.5-2.5μm. Two visibility curves are presented according to the direction, upwind or downwind, for which the observation was made. Figure 1 also includes the calculated Koschmieder scattering (extinction) coefficient for visible light, b_{scat}, a qualitative measurement of visibility change. Visibility reductions near 50% of upwind values are noted to occur between 2-3 hours downwind in agreement with other St. Louis studies (Shea and Auer, 1976; White et al., 1976). Increases over regional upwind values (930 cm^{-3}) in the number of CCN activated in the broad general downwind plume of St. Louis are seen to peak at 77% at about 3 hours travel time. Similiar increases of up to 200% have been observed 2.5 hours downwind of Buffalo (Kocmond and Mack, 1972) and near 300% at 4 hours in a long narrow plume emanating from a paper mill in Washington (Eagan et al., 1974). In the St. Louis plume, the number of 2μm aerosols does not maximize until 2 hours, although there is an earlier, smaller peak in the number of 0.5 hour downwind, presumably reflecting a primary production mechanism {e.g., vehicle exhaust (Habibi, 1973)} for these larger aerosols. Concentrations of aerosols near 0.5μm reach their highest values at 3 hours. Since the CCN samples were collected on the inbound leg, the agreement in time between the inbound visibility minimum and the peak in the CCN and 0.5μm aerosol concentrations

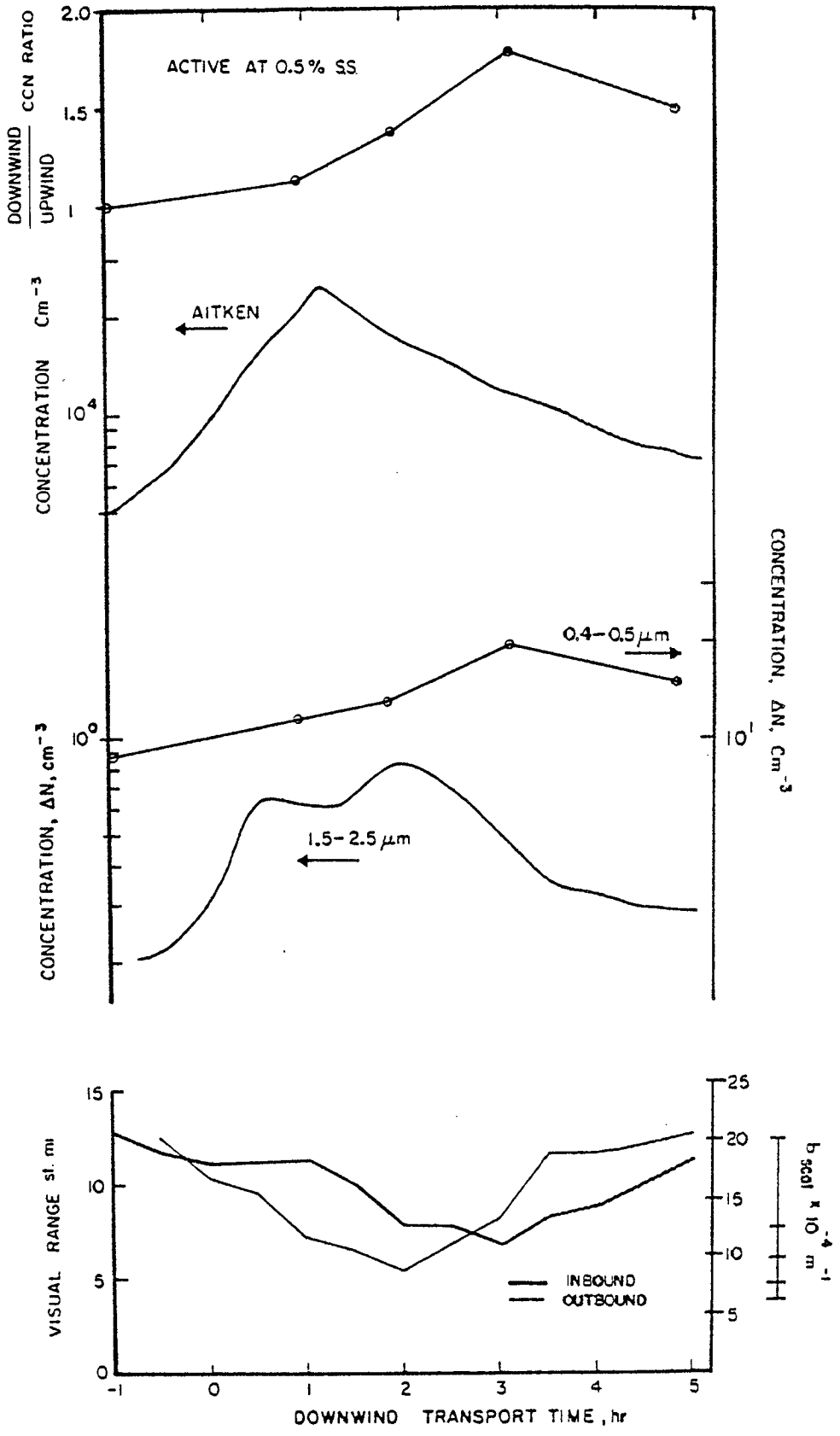

Figure 1: Time section of visual range with corresponding CCN ratio, Aitken nucleus concentration, and numbers of particles in the diameter range 0.4-0.5μm and 1.5-2.5μm observed downwind of St. Louis.

is comforting. Aitken nucleus concentrations are seen to increase above regional background values of 5000 cm^{-3} by fivefold within one hour downwind in agreement both in magnitude and time from recent St. Louis studies (Auer, 1975; Shea and Auer, 1976). The times of lowest visibilities do not coincide with periods of maximum Aitken nucleus concentrations, suggesting that the portion of the aerosol spectra represented by the numbers of 0.5μm and 2.0μm particles (rather than the total number of aerosols) are responsible for the visibility reduction (Knoll et al., 1968).

Thus, in summary from Figure 1, it is seen that a reduction of nearly 50% in the visual range occurs 2-3 hours downwind of the St. Louis Metropolitan area; this visibility restriction appears positively correlated in time with increases in the number of certain optically significant sized aerosols.

The evolution of the full range of particle sizes between 0.02μm and 2.5μm for aerosols passing over the downwind of St. Louis is shown in Figures 2-4. Comparing the particle size disbributions (Figure 2) one hour upwind of the city with those at various times downwind, it can be seen that aerosols less than 0.035μm diameter are present in concentrations significantly higher at all times downwind, maximizing at about 1 hour. The highest concentration of aerosols of this size (Aitken nuclei) at about 1 hour downwind reflects greater numbers of particles by primary production due to anthropogenic sources (e.g., vehicle exhaust and combustion) within the metropolitan area. The concentrations of aerosols between 0.05-1.0μm, representing the range of the light-scattering particles, are seen to increase through 3-hour transport time before falling off slightly at about 5 hours to values nearly equal to those about 2 hours downwind. For sizes 1.5-2.5μm, the greatest numbers are seen 1-2 hours downwind; concentrations at 5 hours are beginning to return to regional upwind values.

An estimate of the complete size spectra permits the calculation of the surface and volume moments of the spectra as well as the total number concentration. The evaluation of the development, as a function of time downwind, of the aerosol volume fraction increases continuously with time in spite of the decrease of the total number of concentration.

Figure 3 shows the surface area-diameter distributions and their change downwind of the city. The total aerosol surface area between 0.05-1.0μm is seen to increase above regional background values up to about 3 hours downwind.

Indicated in Figure 4 are two volume spectra at one hour upwind and 3 hours downwind where aerosol growth by atmospheric processes generating condensable material was active. Within 3 hours travel time of the city, a significantaerosol volume production was observed which took place in the 0.07-2.5μm range, maximizing between 0.8-1.0μm. The aerosol growth is believed to maximize near 3 hours since a later volume distribution at 5 hours shows reduced values.

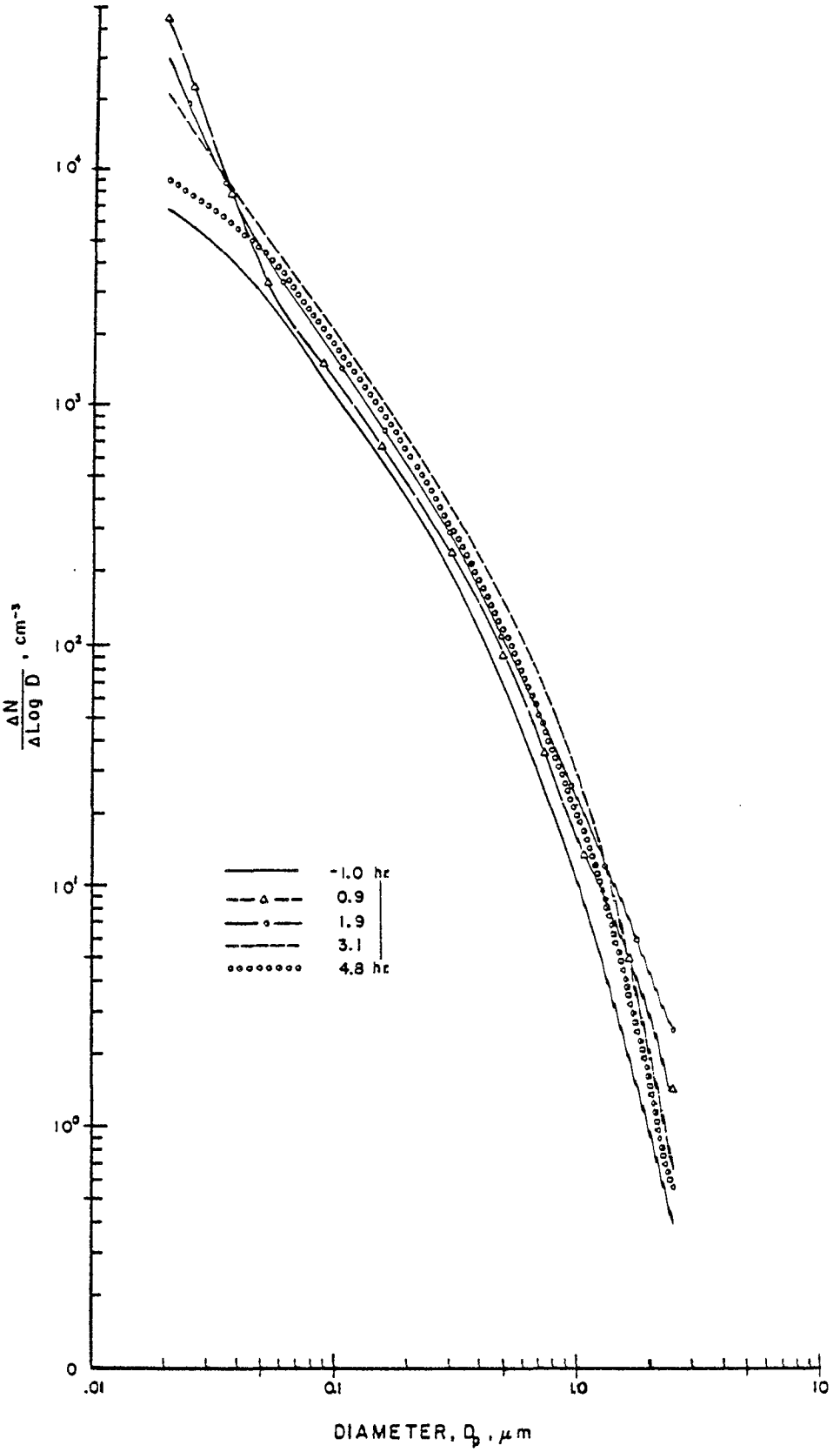

Figure 2: Evolution of the number-size distribution for aerosols downwind of St. Louis.

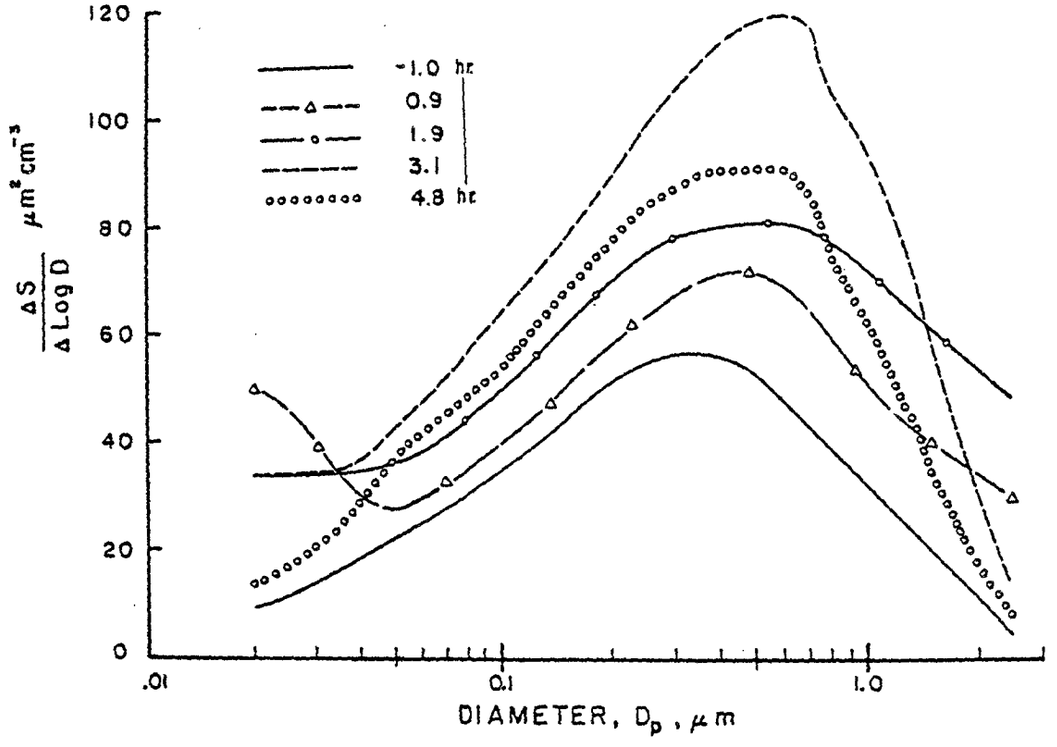

Figure 3: Evolusion of the surface-size distribution of aerosols downwind of St. Louis.

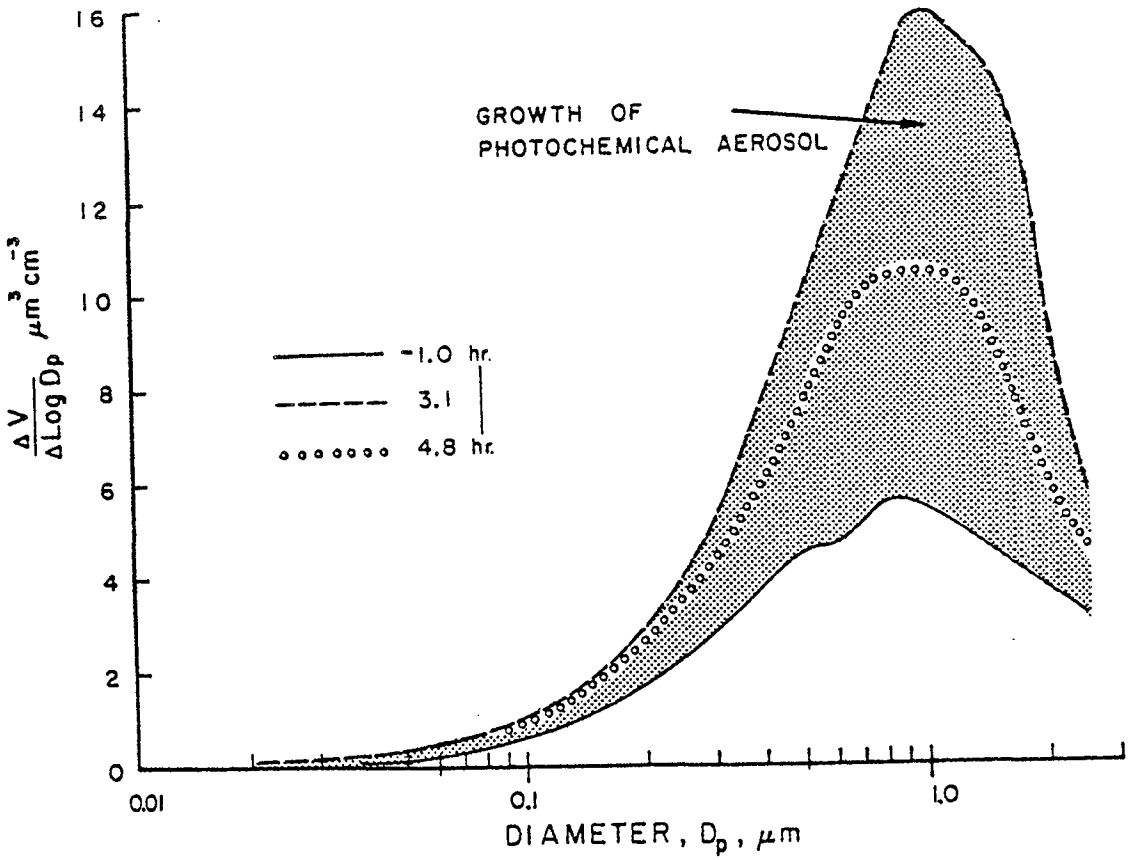

Figure 4: Evolution of the volume-size distribution for aerosols downwind of St. Louis.

Note the constancy of the size at which the mode is located, 0.8-1.0μm. The descriptive characteristics of our volume distribution (first mode peak and saddle point) seem to be shifted toward slightly larger diameters, by a few tenth micrometer (0.9μm versus 0.3μm), than those reported for Los Angeles and Minneapolis (Whitby et al., 1972) and Denver (Durham et al., 1975). However, other reported number distributions for St. Louis (Graham, 1972; Alkezweeny, 1975) yield collaborative volume distributions with the mode at diameters ≥ 0.8-1.0μm.

Information gleaned from Figures 2-4 was used to construct Figure 5, illustrating the evolution of the total number concentration, total surface area, and total aerosol volumes for times downwind. The evolution of photochemical aerosols may be observed by comparing the total surface area and total volume fraction. The volume fraction increases continuously with time to 2-3 hours in spite of the decrease of the total number of concentrations, agreeing qualitatively with a similar presentation (Husar and Whitby, 1973) for a laboratory study of photochemical aerosol formation.

Furthermore, these laboratory studies (Husar and Whitby, 1973) allowed the determination of experimental values for equilibrium surface area at various aerosol formation rates, dV/dt. Thus, a qualitative criterium is established for determining the nature of photochemical gas-particle conversion in a heterogeneous system, the system being defined by the total aerosol surface area, S, and by volumetric conversion rate, dV/dt. For the St. Louis data in Figure 5, the relationship (Husar and Whitby, 1973) suggests that the atmosphere upwind of St. Louis may be just capable at times in providing foreign nuclei with surface area more than sufficient to accomodate photochemical reaction products; i.e., heterogeneous nucleation. However, since a considerable fraction of smog aerosol in the submicron range is hygroscopic and thus sensitive to increasing values of realtive humidity in the mixing layer (Husar et al., 1972), there are aerosols with size, surface, and volume distributions such as those observed in St. Louis (Alkezweeny, 1975) in relative humidities exceeding 80%[2] which are sufficient to allow heterogeneous nucleation to proceed. Declining visibility in high mixing layer relative humidity is an observation easily documented by flight experience at St. Louis.

CONCLUSIONS

A visibility reduction amounting to 50% of prevailing regional upwind visibilities has been found to occur at transport times of 2-3 hours downwind of the St. Louis Metropolitan area. This visibility minimum corresponds in space and time to increased values of CCN active at 0.5% supersaturation. Aerosol distributions, derived from the CCN spectra and in situ optical counting devices, indicate that increases in the number of particles in the 0.1-2.5μm diameter range are responsible

[2]While Alkezweeny did not indicate relative humidity values, the UW team conducted simultaneous flight operations and thus documented prevalent meteorological conditions.

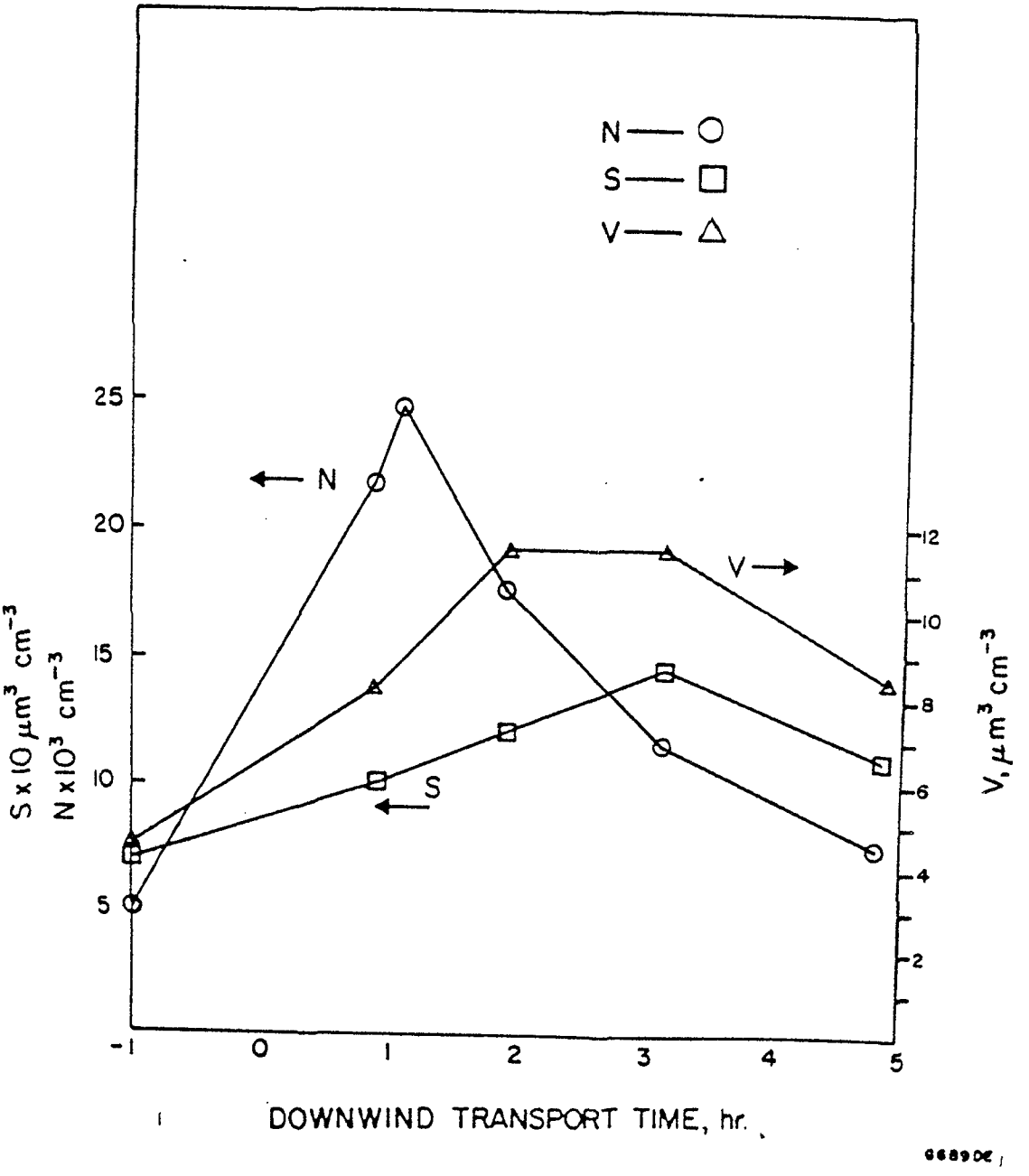

Figure 5: Evolution of aerosol parameters N (total number concentration), S (total aerosol surface area) and V (total aerosol volume fraction) downwind of St. Louis.

for the reduction in visibility. Furthermore, a comparison of the evolution of the aerosol distribution for size, surface area and volume with those derived from laboratory studies suggest that most of the light scattering aerosols are of secondary origin, produced by heterogeneous gas to particle conversions. The secondary conversion products tend to accumulate in the particle size range between 0.1-2.5μm, maximizing around 0.8-1.0μm within 2-3 hours because of some form of the growth law and the shape of the primary aerosol size distribution. This elevated size range may be explained by the fact that St. Louis is located in a region of the country in which large SO_2 sources (mainly power plants, non-ferrous metal smelters and oil refineries) are spaced from a few tens to a few hundreds of kilometers apart in virtually all directions extending for several hundred kilometers. Since the typical removal distance for SO_2 and its oxidation products is probably on the order of a thousand kilometers or more (Rodhe et al., 1972), any site in the region will constantly be under the influence of the SO_2 oxidation products from a large number and a variety of sources, thus providing a unique regional background aerosol distribution (Charlson et al., 1974).

Additional work is planned to further document the extent of the visibility minimum downwind of the urban area and to provide more detail of the aerosol morphology relating to the effects of the humidity, source regions and chemical composition.

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