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D. A. Lundgren & D. W. Cooper

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D. A. Lundgren and  
D. W. Cooper  
Pennsylvania State University

# Effect of Humidity On Light-Scattering Methods Of Measuring Particle Concentration

An experimental investigation was undertaken to isolate and quantitatively determine the effect relative humidity has on the light-scattering ability of aerosols. Both the naturally-occurring ambient aerosol of State College, Pa., and several common test aerosols were used. A measured flow of aerosol was mixed with a measured flow of particle-free air to form a mixture of constant contaminant level; the humidity of this mixture was varied by controlling the moisture content of the clean diluent air. The total light scattered by a given aerosol sample, at various relative humidities, was measured with a Sinclair-Phoenix aerosol photometer (measures the total light scattered in the near forward direction). All measurements were carried out at atmospheric pressure, and after the particulates had an average of 1½ minutes to reach equilibrium with the water vapor. Natural and laboratory-generated aerosols were both tested in this manner.

Light scattering is the basis of several devices used both in the laboratory and in the field to estimate particle concentration. The purpose of this study was to isolate and to determine quantitatively the effect relative humidity has on the total light-scattering method of estimating particulate concentration. Naturally-occurring ambient aerosols and several common test aerosols were investigated in carefully controlled experiments.

## Light-Scattering Theory

The interaction between light and a small particle is complex. The three basic light-scattering regimes — determined by  $\alpha$ , the ratio of the circumference of the particle to the wavelength of the light ( $\alpha = 2\pi r/\lambda$ ) — are discussed briefly below.

**Rayleigh Scattering:** For gas molecules and Aitken nuclei with diameters less than about  $0.06\mu$  ( $\alpha < 0.5$  for green light), the intensity of light scattered per unit illumination is given by<sup>1</sup>

$$I = \frac{8\pi^4 r^6}{R^2 \lambda^4} \left[ \frac{m^2 - 1}{m^2 + 2} \right]^2 (1 + \cos^2 \theta) \quad (1)$$

where

- $I$  = intensity of light scattered in the direction  $\theta$
- $r$  = particle radius
- $m$  = index of refraction of particle
- $\lambda$  = wavelength of light
- $R$  = distance from particle to observer
- $\theta$  = angle of observation with respect to the forward direction of illumination

This is the well-known  $r^6/\lambda^4$  dependence which, among other things, explains the red color of sunsets. For very small particles, Rayleigh's formula is quite useful.

**Mie Scattering:** As  $\alpha$  becomes greater than 0.5, the scattering pattern loses its symmetry and becomes a very complicated function of  $\alpha$  and  $\theta$ . This function, the Mie scattering function, requires numerical solutions; tables of Mie theory computations are available.<sup>2</sup> Because the function does not change monotonically for monochromatic light and an increase in particle size, it is difficult to use Mie scattering from monochromatic light to measure particle size. However, when the contributions of the scattering functions are added together — that is, when the

illumination is white light — the result is scattering roughly proportional to  $r^2$  (for  $r > 0.1\mu$ ). The index of refraction, especially if it is complex (some absorption), also strongly affects light scattering; therefore, the amount of light scattered by a single particle is a function of both its size and its index of refraction.

**Geometrical Optics:** For the largest particles (radius greater than  $5\mu$ ) geometrical optics plus diffraction effects pertain, with scattering proportional to radius squared.

Calculations show that for the usual atmospheric particle size distributions and for visible light, the particles with radii between 0.2 and  $0.4\mu$  contribute most to light scattering, and those with radii less than  $0.1\mu$  contribute negligibly. In general, then, the total light scattered by an aerosol is that scattered by the particles larger than a tenth micron in radius. The fraction of incident white light scattered is proportional to  $\sum n_i k_i \pi r_i^2$  where  $n_i$  is the number of particles per unit volume of a given size and species,  $r_i$  is the particle radius, and  $k_i$  is a light-scattering factor which depends upon refractive index and angle of observation.

## Visibility vs. Light-Scattering

In the atmosphere, the scattering of light by particles both cuts down the amount of light received from a given

Mr. Lundgren is with the State-wide Air Pollution Research Center, University of California, Riverside, California 92502. Mr. Cooper is associated with the Center for Air Environment Studies, Pennsylvania State University, University Park, Pa. 16802.

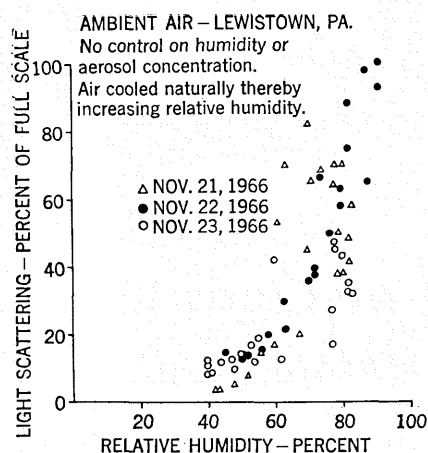


Figure 1. Light scattering vs. humidity in uncontrolled ambient air—Lewistown, Pennsylvania.

object and diminishes the background contrast by diffusing light into areas where there would normally be darkness. (The effect of fog on the light from auto headlights at night is a good example of the way scattering can sharply reduce contrasts and thus, visibility.) Quantitatively, the relation between visibility and scattering is<sup>3</sup>:

$$V = \frac{1}{\sigma} \ln \frac{1}{\epsilon} = \frac{3.91}{\sigma} \quad (2)$$

where

$V$  = visibility in convenient units of length

$\epsilon$  = threshold of perceptible contrast between objects of unequal brightness ( $\epsilon \approx 0.02$ )

$\sigma$  = extinction coefficient in reciprocal of the same units of length as  $V$ ; it is composed of:

$\sigma_m$ , the extinction coefficient of air molecules

$\sigma_n$ , the extinction coefficient of hygroscopic particles and droplets

$\sigma_p$ , the extinction coefficient of non-hygroscopic particles so that  $\sigma = \sigma_m + \sigma_n + \sigma_p$

$\sigma_m$  is small enough that it can be ignored when compared with typical values of  $\sigma_n + \sigma_p$ —for visibilities less than 15 miles.<sup>3</sup>

The extinction coefficient ( $\sigma$ ) can increase due to a change in particle concentration, particle size, or particle index of refraction (composition).

Estimates of the prevailing visibility, even by those accustomed to making them routinely, are obviously subjective. They are also dependent upon the direction in which the observer looks, both because of variations in background and because of variations of the visual properties of air masses with direction (that is, rarely are air

masses homogeneous over distances comparable to those used in estimating visibilities).<sup>6</sup> Such measurements also tell us very little or nothing about ambient contamination during periods of precipitation, when the rain or snow masks the effects of particulates.

#### Visibility vs. Humidity—Review of Past Studies

A relationship between poor visibility and high humidity has been noticed by most people intrepid enough to venture out-of-doors. Thirty years ago, Wright<sup>4</sup> made a quantitative study of the frequency of poor visibility at British weather stations compared with prevailing relative humidity (RH). Buma<sup>5</sup> did much the same thing in Germany, with the added refinement of distinguishing between air masses of continental and maritime origins, noting that both types showed increasing visibility with decreasing humidity, although the continental air showed changes with RH below 70%, whereas the maritime air did not. Similar work was done in the United States by Goldman.<sup>6</sup> He concluded his statistical variations hid any RH-visibility correlation if one existed, a major factor being the variation in estimated visibility with respect to the direction in which it is observed. The major criticism of this kind of study is that voiced by Junge:<sup>7</sup> the correlation may be influenced by a correlation between the origin of the aerosol and the humidity or it could be due to correlations with other meteorological parameters accompanying humidity increases. Thus, to pin down such a correlation, one must be able to change the humidity without changing the other meteorological parameters and without an influx of "foreign" air.

One obvious improvement on the passive correlation technique would be to measure the "contamination" as well as the visibility and the R.H. Hall<sup>8</sup> did this in a study in Los Angeles, and found that, in general, for a given level of particulate contamination (using "reflectance"), the visibility was lower for a higher relative humidity. Two of Hall's criticisms of his work should be noted: the station which supplied the R.H. data was  $\frac{1}{2}$  mile from the station measuring visibility and contamination, and on some days the RH changed by as much as 35% in one hour, the shortest measuring period used. Perhaps more significant is the method used to measure particulate contamination, reflection of light from sampled particles, and the inability of such a method to give much information about photochemical aerosols or liquid droplets.

More recently, Burt<sup>9</sup> ran experiments similar to Hall's in Cincinnati and St. Louis. To measure contami-

nation, he used samplers which drew the air through filter tapes and measured the light transmitted through the tape before and after sampling. As a check, he exposed the soiled tapes to various controlled humidities and found no appreciable change in the readings; this is not surprising, inasmuch as such tape transmission measurements respond essentially only to particle absorption of light, which varies appreciably among species of contaminants, because the filter paper already diffuses the light, be it incident or scattered, thus negating scattering effects. To measure visibility in St. Louis, the principal data in his report, he relied on visual estimates, and to estimate the relevant relative humidity, he relied on dew point measurements made fourteen miles away and corrected with local dry bulb temperature measurements. He found that visibility and RH were correlated and that visibility and "soiling index" were weakly correlated. (Burt's apparatus did not compensate for fluctuations in transmissivity in the tape itself, a factor which he felt important.) As in Hall's work, droplets which can considerably decrease visibility but contain little or no solids would not be measured by the tape measurements.

The growth of particles of a few hundredths of a micron in diameter (Aitken nuclei) with increasing humidity was studied by Junge.<sup>7</sup> Using mobility measurements, he found a definite increase in size with humidity though it was less than would be expected from relative humidity and visibility measurements made by others, his value predicting a decrease in visibility of a factor of 2.2 in going

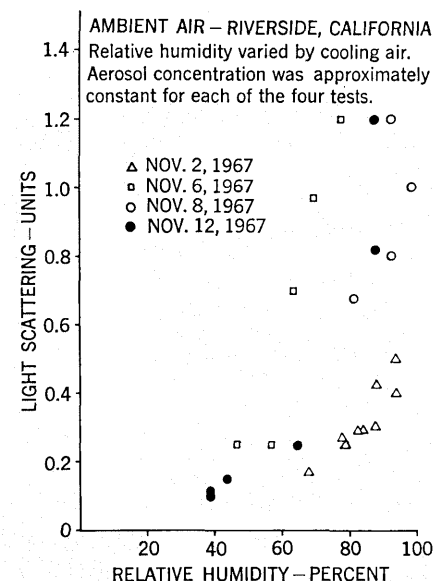


Figure 2. Light scattering vs. humidity in uncontrolled ambient air—Riverside, California.

from 60 to 95% RH. One factor to be considered in assessing this work is that the particles which play a major part in light-scattering visibility changes in the atmosphere are those with radius between  $0.2$  and  $0.4\mu$ , rather than those with radii on the order of hundredths of microns.

Naturally-occurring aerosols present problems: neither their composition nor concentration nor size distribution stay the same for very long; therefore, it is easier and sometimes more valuable to study artificially produced aerosols about which more is known *a priori* and from which, frequently, more can be deduced. Orr<sup>10</sup> and his co-workers studied theoretically and experimentally the behavior of droplets up to  $0.2\mu$  diameter made from various hygroscopic solutions and found generally very good agreement with theory. They showed that not only do these particles increase in size with humidity once they have become droplets, but also that they change phase at different humidities depending upon whether the humidity is increasing or decreasing (hysteresis) and, in addition, that crystals a few hundredths of a micron in diameter become droplets (as R.H. rises) at lower humidities than larger crystals of the same species (e.g., a NaCl crystal  $0.04\mu$  on an edge changes from solid to liquid at 68% RH; the same change for  $1\mu$  and larger NaCl particles occurs at 75% RH). Atmospheric aerosols, composed of a mixture of soluble and insoluble particles and droplets (which may be formed from dissolved gases), cannot be relied upon to behave like artificial aerosols, though the latter give us much useful information.

The possibility of dissolved gases forming aerosol droplets has been mentioned. Renzetti and Doyle<sup>11</sup> produced aerosols from  $0.05$  ppm  $\text{SO}_2$  and 50% RH in air after irradiation with light approximating sunlight. No such aerosol was formed when the irradi-

ation was omitted. Tables of equilibrium concentrations of  $\text{H}_2\text{SO}_4$  in water have been computed as has the expected effect on visibility as a function of humidity.<sup>12</sup>

The data of Figures 1 and 2 show an obvious relationship between light scattering and relative humidity but the data does not provide a measure of the relative air dirtiness independent of humidity. Lewistown, Pa., is a small, somewhat isolated, industrial community with several air polluting industries and serves to represent an industrially polluted area. Riverside, Calif., is a larger community on the east side of the Los Angeles air basin and represents photochemical or automobile related air pollution.

To recapitulate, light scattering is a very useful tool to employ in measuring particulate air pollution. Light scattering, thus visibility, is related to both the amount of aerosol contamination present and the relative humidity of the air. Past studies have not rigorously proved this relationship because the experiments which used naturally-occurring aerosols did not isolate the effect of humidity and those experiments which did isolate the effect of humidity were done only with an "artificial" aerosol.

#### Experimental Procedure

Determining the effect humidity has on light scattering involved dilution of a known flow of aerosol with a known flow of clean, humidified air and then passing this mixture, after equilibration, through a Sinclair-Phoenix aerosol photometer which measured the forward light scattering. Any change in the light scattering was monitored as were the air temperature, flow rate and final relative humidity. A Hygro-dynamics hygrometer of the LiCl type was used to measure RH with an accuracy of  $\pm 2\%$ . A schematic of the experimental apparatus is shown in Figure 3.

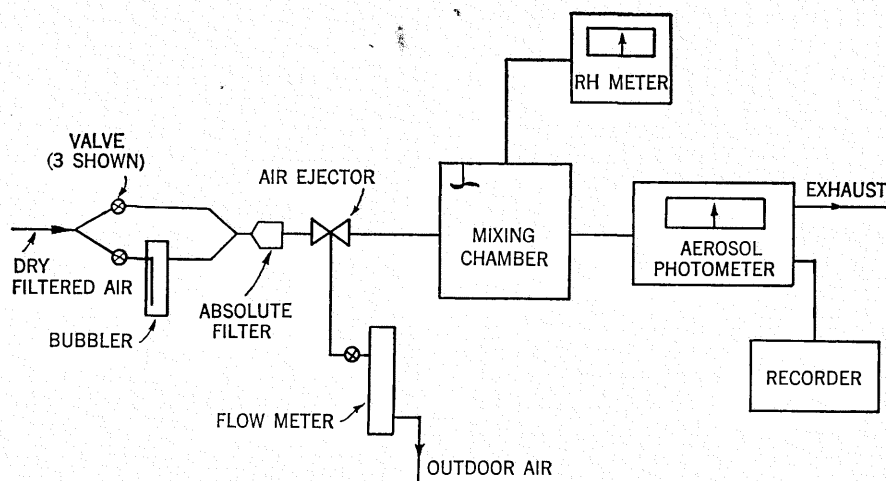


Figure 3. Schematic of experimental setup.

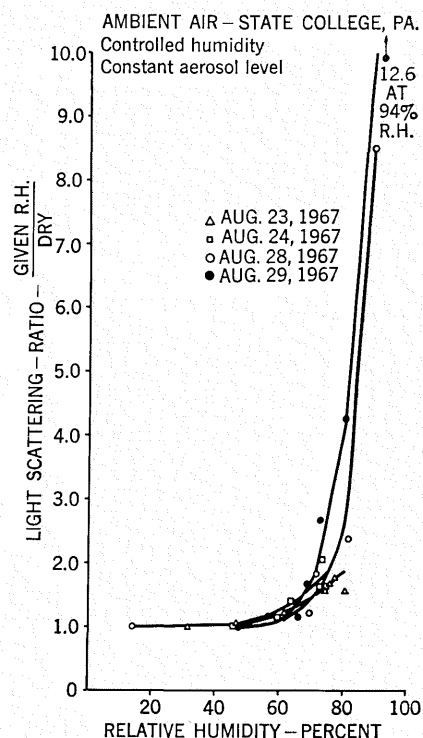


Figure 4. Effect of humidity on light scattering by ambient aerosols.

During operation, a pressurized, filtered airstream was used to "pump" the test aerosol into the system by means of an air ejector—while providing mixing and aerosol dilution at the same time. Various degrees of humidification were achieved by splitting the pressurized air and passing part of it through a fritted glass bubbler immersed in filtered distilled water. After the streams were recombined they were again passed through an absolute filter and then into the air ejector. Dilution air flow of  $0.54$  cfm and aerosol flow of  $0.06$  cfm were used to provide a constant 10 to 1 aerosol dilution ratio. The  $1\text{ ft}^3$  stainless mixing chamber provided ample time for the aerosol and dilution air to mix and reach an equilibrium.

A slight mixing chamber pressure (1 in.  $\text{H}_2\text{O}$ ) prevented any influx of contaminated air. Temperature of the airstreams, mixing chamber, and photometer optical chamber was controlled to assure accurate relative humidity measurements. Several tests were run to assure that the observed changes in light scattering were humidity effects and not contaminants in the dilution-humidification air.

Tests on ambient air were run on several different days; the test results are presented in Figure 4. For better comparison (and because the absolute level of particulates was different on different days) the results are graphed

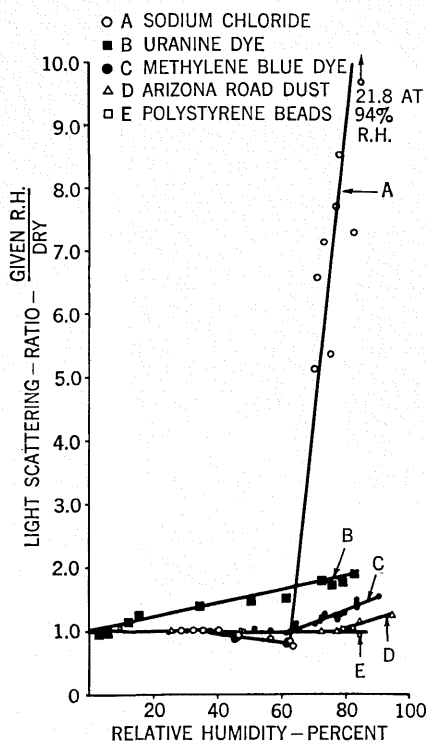


Figure 5. Effect of humidity on light scattered by laboratory aerosols.

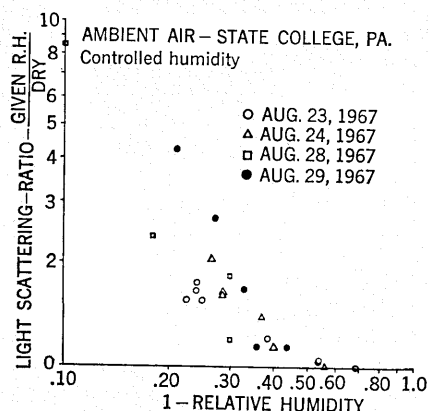


Figure 6. Light scattering vs 1—relative humidity for ambient aerosols.

as the ratio of light scattered at a given relative humidity to that scattered at 15% RH as a function of RH. Figure 4 results agree with other ambient air results obtained several months earlier when the temperatures were not as carefully controlled. The only difference is a greater scatter in the earlier unplotted data.

Test results for several common laboratory test aerosols are presented in Figure 5. For all but the polystyrene beads, the aerosols were produced by atomizing a 1% by volume aqueous solution of the aerosol material with a standard Collison atomizer-ion generator setup described elsewhere.<sup>13</sup> The polystyrene bead aerosol was formed by atomizing about a 0.01% suspension of beads in distilled water.

For all data test points, the light-scattering readings were measured at least 7 min (four mixing chamber air changes) after any change in the test system to assure a dynamic equilibrium. Generally, each measurement at a given humidity was followed by a measurement at a reference humidity of 15% to be sure the absolute aerosol level had not changed.

An operational problem was caused by a heating of the aerosol as it passed through the Sinclair-Phoenix optical chamber. An average temperature rise of 0.5°C in the airstream was measured; for example, this heating effect would lower the relative humidity of a 60% RH airstream by about 2% RH.

Assuming half of this heating effect took place by the time the airstream passed through the optical sensing zone in this case, a 1% RH correction would have been applied. This heating took place even though the optical chamber outer surfaces were cooled somewhat and may further affect the basic  $\pm 2\%$  RH measurement uncertainty.

#### Discussion of Results

Figures 1 and 2 suggest a rather strong relationship between relative humidity and light scattering, both for aerosols which are mainly created by industry (Lewistown, Pa.) and for those which are of a photochemical nature (Riverside, Calif.). The data in both cases also clearly illustrates a strong day-to-day fluctuation (at any given RH) in the absolute "dirtiness" of the air. A difference in slope of the daily results can also be noted, although it is less apparent.

To determine quantitatively the effect of humidity on light scattering by a given ambient aerosol, the data presented in Figure 4 was obtained. Although data for only four different air masses (listed by date) are plotted, the result is rather obvious and dramatic. Results are plotted as a ratio of the scattering at a given relative humidity to that scattered at 15% RH (noted as "dry" on the figure). Results for the four air masses are therefore directly comparable although the absolute aerosol concentrations were

different. Data scatter at 80% RH illustrates a definite difference in the four air masses. This difference is more obvious in Figure 6 which is a logarithmic plot of the scattering ratio vs. 1—RH (for this same data). A best fit line can be drawn through each day's data. The difference in the slope of these lines is very important in that it shows that no single correction factor can be applied to allow direct comparison of two different high humidity light-scattering measurements made on two different aerosols. For example, the aerosol sample of August 29 had a fourfold change in light scattering in going from 50 to 80% RH, while the aerosol sample of August 23 had less than a twofold change. However, this data and earlier data<sup>14</sup> indicate that any light-scattering measurements made at humidities below 30 or 40% are directly comparable; therefore, if any two different aerosol samples are both brought to this condition of low RH (by heating for example) they may be directly compared. The hysteresis effects shown by Orr, *et al.*,<sup>10</sup> were not specifically studied but will be minimal at low (<30%) RH values. If the size distributions of two dry aerosols are similar, the mass loading of the samples will be in about the same ratio as the light-scattering readings.

Differences in the slope of the lines drawn through the data points for the different air masses may be interpreted

as a difference in the hygroscopic or water-soluble fraction of the various air masses. Data shown in Figure 2 can be interpreted in this same way. Actual measurements of the water-soluble fraction of Hi-Vol particulate samples show a range from about 20 to 60% soluble for Riverside, Calif. (water-soluble fraction was not determined for the data of Figure 4). Chemical analysis of typical aerosol samples for both State College and Riverside has shown that a large fraction of the dry particulate weight is crystalline, but of this only a small fraction is sodium chloride.

A comparison of the light-scattering ratio at say 80 to 20% RH may be a useful technique for gaining information about the aerosol composition (i.e., percent water soluble). The humidity at which light scattering begins to increase may also provide useful information. When dealing with droplets of sulfuric acid or other dissolved gases, a continuous rise in light scattering with relative humidity would be expected,<sup>12</sup> rather than a rise beginning at 50% RH or so.

Data for laboratory generated aerosols such as uranine dye, methylene blue dye and sodium chloride show to what extent and when humidity must be a consideration in their use. The indicated uniform behavior of methylene blue aerosols at all relative humidities under 50 or 60% has been observed by the authors and several other investigators. The indicated non-uniform behavior of uranine has also been noted. Water soluble uranine dye particles become liquid droplets at some point below 50% RH and upon later drying may (and often do) form particles of non-spherical or crystalline shape. At 80% RH the uranine aerosol scattered 1.8 times as much light as it did at 5% RH. Methylene blue, however, did not change until a relative humidity of 60%, and at 80% RH the light scattering had only increased by a factor of 1.3.

Data for the sodium chloride aerosol rather dramatically illustrates what

can happen with a hygroscopic material. The experimental results showing a rapid change in light scattering at about 70% are in accord with the findings of Orr, *et al.*<sup>10</sup> A very interesting dip in the scattering curve occurred before the phase change. This was noted early in the study and was repeated later under "optimum" conditions with the same results being obtained. Experimental checks and calculations could not account for the apparent "loss" by either sedimentation or coagulation. Orr, *et al.*,<sup>10</sup> noted that at 30% RH there is a shell of water a few molecules thick adsorbed on the crystalline particle, and the effect of this shell may be to lower the scattering in certain directions.

Much research and calibration of light-scattering devices are done using polystyrene latex beads; therefore, it was useful to find no apparent increase in light scattering with relative humidity for 0.36 $\mu$  diameter beads. Tests with beads were run at humidities up to 85%.

Arizona Road Dust is a relatively non-hygroscopic dust often used to test commercial filters. No changes in light scattering were observed until the relative humidity exceeded 80%; the small increase was probably due to the small hygroscopic fraction of the dust.

### Conclusions

Humidity should be taken into account when making light-scattering measurements or visibility determinations. The light scattered by naturally-occurring aerosols may start to increase at relative humidities of 50% or less, depending upon the aerosol hygroscopic fraction. Aerosol samples can best be compared by light scattering if they are dry or are dried to a reasonably low relative humidity, perhaps below 30%. Light-scattering particle counters are also affected by humidity and will count and size hygroscopic particles differently at different humidities. The ratio of light scattered at 2 RH values can provide useful information about the water-

soluble constituents present in that aerosol. Laboratory test aerosols such as methylene blue and uranine should be used with an awareness that their mean particle size is humidity dependent.

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