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Models for the Aerosols of the Lower Atmosphere and the Effects of Humidity Variations on Their Optical Properties

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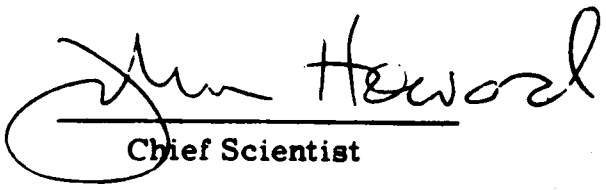

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Aerosol models have been developed for the lower atmosphere. These models are representative of conditions found in rural, urban, and maritime air masses. The changes in the aerosol properties with variations in the relative humidity are discussed. To describe the aerosol optical properties in the extreme of 100 percent relative humidity, several fog models are presented. For each model the coefficients for extinction, scattering, and absorption, the angular scattering distribution, and other optical parameters have been computed for wavelengths between 0.2 and 40 μm. These aerosol models are		

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presented together with a review of their experimental basis. The optical properties of these models are discussed and some comparisons of the model with experimental measurements are presented.

Preface

We would like to thank several individuals who helped with this report, in particular: Fred Volz for his advice on the aerosol refractive indices and his general comments on the aerosol models, Frank Gibson for his work in developing the Fog Models, and Barry Siegel for his assistance with the computer programming.

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Models for the Aerosols of the Lower Atmosphere and the Effects of Humidity Variations on Their Optical Properties

1. INTRODUCTION

Propagation of electromagnetic radiation at optical/infrared frequencies through the atmosphere is affected by absorption and scattering by air molecules and by particulate matter (haze, dust, fog, and cloud droplets) suspended in the air. Scattering and absorption by haze particles or aerosols becomes the dominant factor in the boundary layer near the earth's surface, especially under low visibility conditions.

Atmospheric aerosol particles in the atmosphere vary greatly in their concentration, size, and composition, and consequently in their effects on optical and IR radiation.

There are many scientific and technical reasons why it is necessary to develop models for atmospheric aerosols. They are needed to make estimates of the transmittance, of angular light scattering distribution, of contrast reduction, sky radiance, or other atmospheric optical properties or effects.

Models for the optical properties of aerosols have been developed previously at AFGL and elsewhere.[1-7*] For the lower layer near the earth's surface, these models define an average continental type aerosol whose concentration can be scaled according to surface visibility.

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*Due to the large number of references cited in this report, they will not be footnoted. See References, pages 89 through 94.

The aerosol properties in these models were based on experimental measurements that were made during and prior to the mid-1960's. At that time there was sufficient experimental data available to define an average aerosol model with some different haze concentrations in the lower troposphere (up to a few km altitude) with exponential vertical decrease in particle concentration.

During the past decade, in this country and elsewhere, extensive additional measurements from ground as well as airborne platforms have been made of aerosol concentrations, their size distribution, and optical properties, to warrant the development of updated aerosol models that also describe some of the temporal and spatial variations in atmospheric aerosol distributions and properties. There are now sufficient experimental data to develop models for several different types of tropospheric aerosols, including the dependence of the aerosol properties on relative humidity.

Such updated models have been developed by Shettle and Fenn[8] and Toon and Pollack,[9] except both of these sets of models neglect the effects of relative humidity. The present report describes aerosol models for the lower atmosphere and their optical properties including a discussion of how the aerosol properties change as a function of relative humidity. The optical properties of the models are given for a number of wavelengths between 0.2 and 40 μm, and for several different relative humidities ranging from 0 to 99 percent. In addition four fog models are given for the droplet-condensation phase.

The models of the atmospheric aerosols and their optical properties presented below are based on a review of the available data on the nature of the aerosols, their sizes, their distribution, and variability. However, it must be emphasized that these models represent only a simple, generalized version of typical conditions. It is not practical to include all the details of natural aerosol distributions nor are existing experimental data sufficient to describe the frequency of occurrence of the different conditions. While these aerosol models were developed to be as representative as possible of different atmospheric conditions, the following point should be kept in mind when using any such model: Given the natural variability of the atmospheric aerosols almost any aerosol model is supported by some measurements and no model (or set of models) will be consistent with all measurements.

2. MODELS FOR THE PHYSICAL PROPERTIES OF THE AEROSOLS

2.1 Model Size Distribution

The size distributions for the different aerosol models are represented by one or the sum of two log-normal distributions:

$$n(r) = \frac{dN(r)}{dr} = \sum_{i=1}^{2} \left(\frac{N_i}{\ln(10) \cdot r \cdot \sigma_i \sqrt{2\pi}} \right) \exp \left[- \frac{(\log r - \log r_i)^2}{2 \sigma_i^2} \right] * \quad (1)$$

where N(r) is the cumulative number density of particles of radius r; σ is the standard deviation; r_i, N_i are the mode radius and the number density with r_i. This form of distribution function represents the multimodal nature of the atmospheric aerosols that has been discussed in various studies.[10-14] While Harris and McCormick[15] have suggested using the sum of four log-normal distributions and Davies[16] has used the sum of as many as seven log-normal distributions to fit a measured aerosol size distribution, Whitby and Cantrell[17] have shown that two modes are generally adequate to characterize the gross features of most aerosol distributions. While a third component is often necessary to represent the Aitken nuclei especially near sources of combustion particulates, their effect on the optical properties is small and will be neglected.

There are measurements showing the composition of the atmospheric particulates depending on their size,[18, 19] and using a bimodal size distribution offers the possibility of treating the composition of the individual modes separately. However, there is, in general, insufficient experimental data to uniquely define different refractive index models for the different size ranges, along with differing dependence on relative humidity.

For the maritime conditions, there is evidence[20] showing that the large particles are almost exclusively of oceanic origin and the smaller particles are predominantly of the same composition as the continental aerosols so that we do not differentiate between the two size ranges in terms of their composition.

Four different aerosol models for the atmospheric boundary layer near the earth's surface have been developed. They differ in particle size distribution and particle refractive index. Table 1 lists the parameters defining the size distributions in accordance with Eq. (1) for these models.

The choices of N in Table 1 are normalized to correspond to 1 particle/cm^3. The actual size distributions can be re-normalized to give the correct extinction coefficients for the altitude and for the visibility being used. The continental and oceanic components of the maritime model can be used in various proportions depending on the prevailing winds—particularly in coastal regions. The basis for the characterization of each of the aerosol models is discussed in Sections 2.3 through 2.6.

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*Following the usual convention, log is the logarithm to the base 10 and ln is the logarithm to the base e.

Table 1. Characteristics of the Aerosol Models of the Lower Atmosphere

Aerosol Model	Size Distribution			Type
	N_i	r_i^*	σ_i	
RURAL	0.999875 0.000125	0.03 0.5	0.35 0.4	Mixture of Water-Soluble and Dust-Like Aerosols
URBAN	0.999875 0.000125	0.03 0.5	0.35 0.4	Rural Aerosol Mixture with Soot-Like Aerosols
MARITIME				
Continental Origin	1.	0.03	0.35	Rural Aerosol Mixture
Oceanic Origin	1.	0.3	0.4	Sea Salt Solution in Water
TROPOSPHERIC	1.	0.03	0.35	Rural Aerosol Mixture

*These mode radii correspond to moderate humidities (70 to 80%); values of r_i as function of humidity are given in Table 2.

Table 2. Mode Radii for the Aerosol Models as a Function of Relative Humidity

Relative Humidity	Tropospheric	Rural		Maritime	Urban	
	r_1	r_1	r_2		r_1	r_2
0%	0.02700	0.02700	0.4300	0.1600	0.02500	0.4000
50%	0.02748	0.02748	0.4377	0.1711	0.02563	0.4113
70%	0.02846	0.02846	0.4571	0.2041	0.02911	0.4777
80%	0.03274	0.03274	0.5477	0.3180	0.03514	0.5805
90%	0.03884	0.03884	0.6462	0.3803	0.04187	0.7061
95%	0.04238	0.04238	0.7078	0.4606	0.04904	0.8634
98%	0.04751	0.04751	0.9728	0.6024	0.05996	1.1691
99%	0.05215	0.05215	1.1755	0.7505	0.06847	1.4858

2.2 Effects of Humidity Variations on Aerosol Properties

As the relative humidity increases, water vapor condenses out of the atmosphere onto the particulates suspended in the atmosphere. This condensed water increases the size of the aerosols and changes their composition and their effective refractive index. The resulting effect of the aerosols on the absorption and scattering of light will correspondingly be modified. There have been a number of studies of the change of aerosol properties as a function of relative humidity.[21-28] The most comprehensive of these, especially in terms of the resulting effects on the aerosol optical properties is the work of Hänel.[26-28]

The change in the particulate size is related to the relative humidity by (following Hänel's notation)

$$r(a_w) = r_o \left[1 + \rho \cdot \frac{m_w(a_w)}{m_o} \right]^{1/3} \tag{2}$$

where

r_o is the dry particle radius,

ρ is the particle density relative to that of water,

$m_w(a_w)$ is the mass of condensed water,

m_o is the dry particle mass, and

a_w is the water activity which is essentially the relative humidity f, corrected for curvature of the particle surface.

$$a_w = f \cdot \exp\left(\frac{-2\sigma V_w}{R_w \cdot T \cdot r} \right) \tag{3}$$

where

σ = surface tension on the wet particle surface,

V_w = specific volume of water,

R_w = specific gas constant for water,

T = absolute temperature (°K).

For room temperature (T = 298°K),

$$\frac{2\sigma V_w}{R_w T} \approx 0.001056 \text{ [micron]} \text{ (Hänel,}^{28} \text{ page 126)}.$$

Typical atmospheric temperatures are as much as 20 percent lower but, for particle radii $r > 0.01\ \mu m$, this leads to errors of less than 2 percent in curvature effect so Eq. (3) can be rewritten as

$$a_w = f \cdot \exp\left(\frac{-0.001056}{r(a_w)} \right), \tag{4}$$

where r is in μm and where the dependence of r on a_w has been made explicit.

There are a number of studies on change in size or mass of aerosol particles as a function of relative humidity for various electrolytes[21, 25] and natural atmospheric particulates.[11, 12, 15, 17, 18, 19, 22, 23, 26, 28, 29, 30] Hänel[28] (in his Table IV) has tabulated his and other measurements of $\frac{m_w(a_w)}{m_o}$ vs a_w for various types of

natural aerosols. However, even with this data on the relative mass of condensed water for use in Eq. (2), it is not possible to combine Eq. (2) and (4) into an exact analytic expression giving aerosol radius, r, as an explicit function of relative humidity, because a_w appears on both sides of Eq. (4). Various approximations have been developed.[25, 28, 31] However, these tend to breakdown for small particle sizes and high humidities.

To avoid the limitations of these approximations, Eq. (2) and (4) were used alternately in an iterative manner until they converged (typically 5 or 6 iterations) starting with $a_w \approx f$ on the right side of Eq. (2). Starting with $r \approx r_o$ in Eq. (4) leads to the same result. To interpolate between Hänel's[28] data for different water activities, a_w^i and a_w^{i+1}, it was assumed that

$$\frac{m_w(a_w^i)}{m_w(a_w^{i+1})} \approx \left(\frac{1-a_w^i}{1-a_w^{i+1}} \right)^{\nu_i} . \tag{5}$$

Once the wet aerosol particle size is found from Eq. (2) and (4), the effective complex refractive index, n, is simply the volume weighted average of the refractive indexes of the dry aerosol substance, n_o, and water, n_w. Equivalently, this can be written as

$$n = n_w + (n_o - n_w) \cdot \left[\frac{r_o}{r(a_w)} \right]^3 . \tag{6}$$

For the refractive index of water, the survey of Hale and Querry[32] was used. While there are some minor differences between the optical constants in Hale and Querry's survey and the more recent measurements[33, 34] these differences are comparable with the experimental errors and are small compared with the other uncertainties in the model parameters. These refractive index data are shown in Figure 1.

2.3 Rural Aerosol Model

The "Rural Model" is intended to represent the aerosol under conditions where it is not directly influenced by urban and/or industrial aerosol sources. The rural aerosols are assumed to be composed of a mixture of 70 percent of water soluble substance (ammonium and calcium sulfate and also organic compounds) and 30 percent dust-like aerosols. The refractive index for these components based on the measurements of Volz[35, 36] is shown in Figure 2 and tabulated in Table 3. These refractive index data weighted by the mixing ratio of the two components are consistent with other direct measurements,[37, 38] and with values inferred from in situ measurements.[39-41]

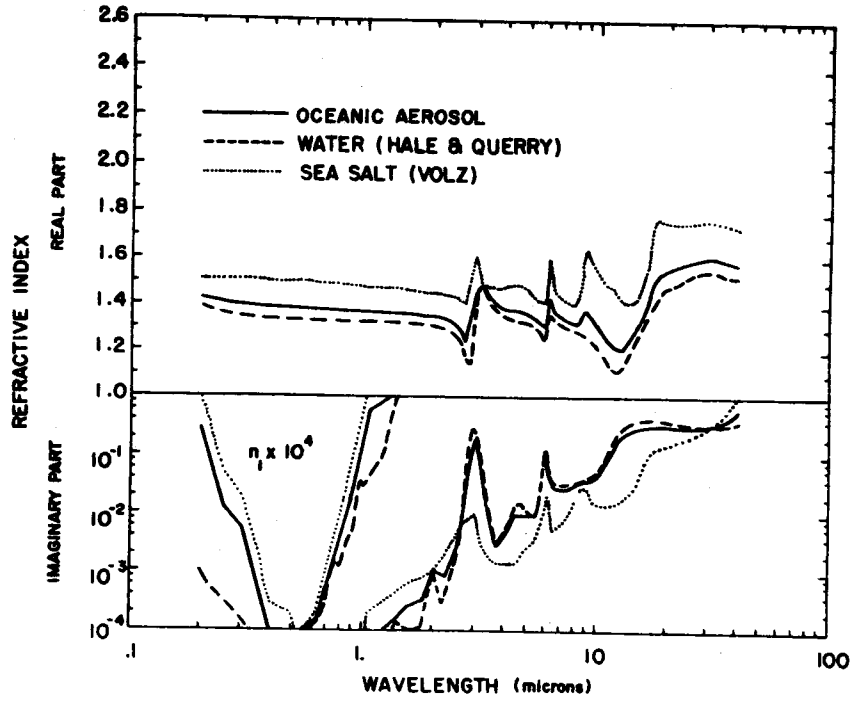

Figure 1. Refractive Index of Oceanic Aerosol, Water, and Sea Salt

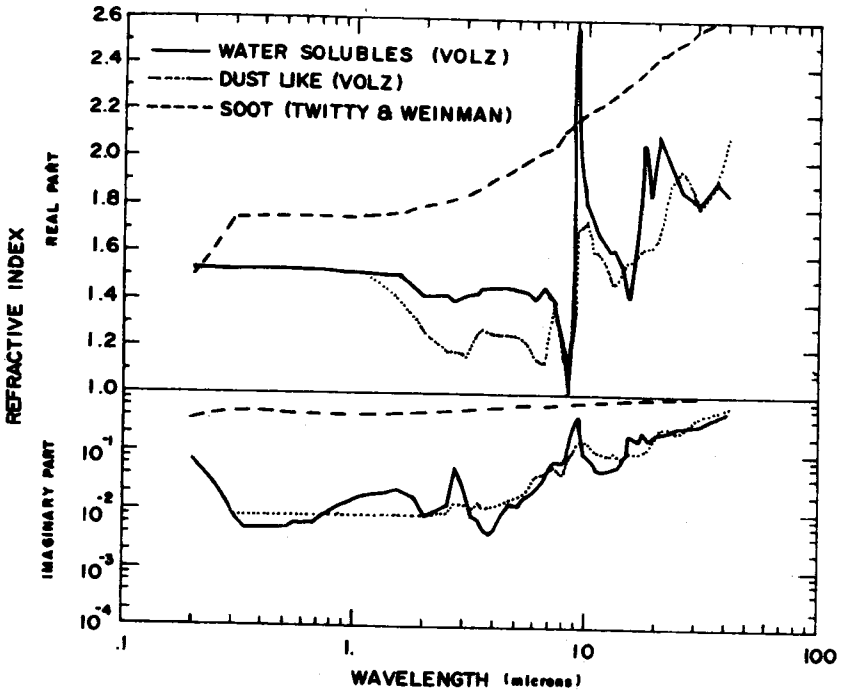

Figure 2. Refractive Index for the Dry Rural and Urban Aerosol Components

Table 3. Refractive Index for the Different Aerosol Components

WAVELENGTH (MICRONS)	WATER SOLUBLE		DUST-LIKE		SOOT		SEA SALT		WATER	
.2000	1.530	-7.00E-02	1.530	-7.00E-02	1.500	-.350	1.510	-1.00E-04	1.396	-1.10E-07
.2500	1.530	-3.00E-02	1.530	-3.00E-02	1.620	-.450	1.510	-5.00E-06	1.362	-3.35E-08
.3000	1.530	-8.00E-03	1.530	-8.00E-03	1.740	-.470	1.510	-2.00E-06	1.349	-1.60E-08
.3371	1.530	-5.00E-03	1.530	-8.00E-03	1.750	-.470	1.510	-4.00E-07	1.345	-8.45E-09
.4000	1.530	-5.00E-03	1.530	-8.00E-03	1.750	-.460	1.500	-3.00E-08	1.339	-1.86E-09
.4880	1.530	-5.00E-03	1.530	-8.00E-03	1.750	-.450	1.500	-2.00E-08	1.335	-9.69E-10
.5145	1.530	-5.00E-03	1.530	-8.00E-03	1.750	-.450	1.500	-1.00E-08	1.334	-1.18E-09
.5500	1.530	-6.00E-03	1.530	-8.00E-03	1.750	-.440	1.500	-1.06E-08	1.333	-1.96E-09
.6328	1.530	-6.00E-03	1.530	-8.00E-03	1.750	-.430	1.490	-2.00E-08	1.332	-1.46E-08
.6943	1.530	-7.00E-03	1.530	-8.00E-03	1.750	-.430	1.490	-1.00E-07	1.331	-3.05E-08
.8600	1.520	-1.20E-02	1.520	-8.00E-03	1.750	-.430	1.480	-3.00E-06	1.329	-3.29E-07
1.0600	1.520	-1.70E-02	1.520	-8.00E-03	1.750	-.440	1.470	-2.30E-04	1.326	-4.18E-06
1.3000	1.510	-2.00E-02	1.460	-8.00E-03	1.760	-.450	1.470	-4.06E-04	1.323	-3.69E-05
1.5360	1.510	-2.30E-02	1.400	-8.00E-03	1.770	-.460	1.460	-6.00E-04	1.318	-9.97E-05
1.8000	1.460	-1.70E-02	1.330	-8.00E-03	1.790	-.480	1.450	-8.00E-04	1.312	-1.15E-04
2.0000	1.423	-8.00E-03	1.260	-8.00E-03	1.800	-.490	1.450	-1.80E-03	1.306	-1.10E-03
2.2500	1.420	-1.00E-02	1.220	-9.00E-03	1.810	-.500	1.440	-2.00E-03	1.292	-3.90E-04
2.5000	1.420	-1.20E-02	1.180	-9.00E-03	1.820	-.510	1.430	-4.00E-03	1.261	-1.74E-03
2.7000	1.400	-5.50E-02	1.180	-1.30E-02	1.830	-.520	1.400	-7.00E-03	1.188	-1.96E-02
3.0000	1.420	-2.20E-02	1.160	-1.20E-02	1.840	-.540	1.610	-1.00E-02	1.371	-.272
3.2000	1.430	-8.00E-03	1.220	-1.00E-02	1.860	-.540	1.490	-3.30E-03	1.478	-9.24E-02
3.3923	1.439	-7.00E-03	1.260	-1.30E-02	1.870	-.550	1.480	-2.00E-03	1.422	-2.04E-02
3.5000	1.450	-5.00E-03	1.280	-1.10E-02	1.880	-.560	1.480	-1.60E-03	1.400	-9.40E-03
3.7500	1.452	-4.00E-03	1.270	-1.10E-02	1.900	-.570	1.470	-1.40E-03	1.369	-3.91E-03
4.0000	1.455	-5.00E-03	1.280	-1.20E-02	1.920	-.580	1.480	-1.40E-03	1.351	-4.61E-03
4.5000	1.460	-1.30E-02	1.260	-1.40E-02	1.940	-.590	1.490	-1.40E-03	1.332	-1.34E-02
5.0000	1.450	-1.20E-02	1.250	-1.60E-02	1.970	-.600	1.470	-2.50E-03	1.325	-1.24E-02
5.5000	1.440	-1.80E-02	1.220	-2.10E-02	1.990	-.610	1.420	-3.60E-03	1.298	-1.16E-02
6.0000	1.410	-2.30E-02	1.150	-3.70E-02	2.020	-.620	1.410	-1.10E-02	1.265	-.107
6.2700	1.430	-2.70E-02	1.140	-3.90E-02	2.030	-.625	1.600	-2.20E-02	1.363	-8.86E-02
6.5000	1.460	-3.30E-02	1.130	-4.20E-02	2.040	-.630	1.460	-5.00E-03	1.339	-3.92E-02
7.2000	1.400	-7.00E-02	1.400	-5.50E-02	2.060	-.650	1.420	-7.00E-03	1.312	-3.21E-02
7.9000	1.200	-6.50E-02	1.150	-4.90E-02	2.120	-.670	1.400	-1.30E-02	1.294	-3.39E-02
8.2000	1.019	-.100	1.130	-7.40E-02	2.130	-.680	1.420	-2.00E-02	1.286	-3.51E-02
8.5000	1.300	-.215	1.300	-9.04E-02	2.150	-.690	1.480	-2.60E-02	1.278	-3.67E-02
8.7000	2.400	-.290	1.400	-.100	2.160	-.690	1.600	-3.00E-02	1.272	-3.79E-02
9.0000	2.560	-.370	1.700	-.140	2.170	-.700	1.650	-2.80E-02	1.262	-3.99E-02
9.2000	2.200	-.420	1.720	-.150	2.180	-.700	1.610	-2.60E-02	1.255	-4.15E-02
9.5000	1.950	-.160	1.730	-.162	2.190	-.710	1.580	-1.80E-02	1.243	-4.44E-02
9.8000	1.870	-9.50E-02	1.740	-.162	2.200	-.715	1.560	-1.60E-02	1.229	-4.79E-02
10.0000	1.820	-9.00E-02	1.750	-.162	2.210	-.720	1.540	-1.50E-02	1.218	-5.08E-02
10.5918	1.760	-7.80E-02	1.620	-.120	2.220	-.730	1.500	-1.40E-02	1.179	-6.74E-02
11.0000	1.720	-5.00E-02	1.620	-.105	2.230	-.730	1.480	-1.40E-02	1.153	-9.68E-02
11.5000	1.670	-4.70E-02	1.590	-.100	2.240	-.740	1.480	-1.40E-02	1.126	-.142
12.5000	1.620	-5.30E-02	1.510	-9.00E-02	2.270	-.750	1.420	-1.60E-02	1.123	-.259
13.0000	1.620	-5.50E-02	1.470	-.100	2.280	-.760	1.410	-1.80E-02	1.146	-.305
14.0000	1.560	-7.30E-02	1.520	-8.50E-02	2.310	-.775	1.410	-2.30E-02	1.210	-.378
14.8000	1.440	-.100	1.570	-.100	2.330	-.790	1.430	-3.00E-02	1.256	-.396
15.0000	1.420	-.200	1.570	-.160	2.330	-.790	1.450	-3.50E-02	1.270	-.402
16.4000	1.758	-.160	1.600	-.100	2.360	-.810	1.560	-9.00E-02	1.346	-.427
17.2000	2.080	-.240	1.630	-.100	2.380	-.820	1.740	-.120	1.386	-.429
18.0000	1.980	-.180	1.640	-.115	2.400	-.825	1.780	-.134	1.423	-.426
18.5000	1.850	-.170	1.640	-.120	2.410	-.830	1.770	-.135	1.443	-.421
20.0000	2.120	-.220	1.680	-.220	2.450	-.850	1.760	-.152	1.480	-.393
21.3000	2.060	-.230	1.770	-.280	2.460	-.860	1.760	-.165	1.491	-.379
22.5000	2.000	-.240	1.900	-.280	2.480	-.870	1.760	-.180	1.506	-.370
25.0000	1.880	-.280	1.970	-.240	2.510	-.890	1.760	-.205	1.531	-.356
27.9000	1.840	-.290	1.890	-.320	2.540	-.910	1.770	-.275	1.549	-.339
30.0000	1.820	-.300	1.800	-.420	2.570	-.930	1.770	-.360	1.551	-.328
35.0000	1.920	-.480	1.900	-.500	2.630	-.970	1.760	-.560	1.532	-.326
40.0000	1.860	-.500	2.100	-.600	2.690	-1.000	1.740	-1.000	1.519	-.305

In the preliminary version of these models,[8] the water-soluble and dust-like components were treated separately and the results of the Mie scattering calculations on individual components were combined. To reduce the calculations for the current models, which now are done as a function of humidity, the individual aerosol particles were considered to be a homogeneous combination of the different types of substance—as many natural aerosols are.[42] The resulting refractive index for the composite rural aerosol is given in Table 3. However, Bergstrom[43] has argued that using such mean refractive indexes in determining the optical properties will result in errors.

However, it should be noted that using this composite refractive index and Shettle and Fenn's[8] rural aerosol size distribution and comparing these results for the scattering and absorption coefficients with those based on separate calculations for the different aerosol types, one finds only a 5 percent difference except for the scattering minimum at 8.2 μm where the difference was 16 percent.

The parameters for the rural model size distribution given in Table 1 fall within what Whitby and Cantrell[17] give as a typical range of values for the accumulation and coarse particle modes.

The resulting number density distribution, $n_{(r)}$, is shown in Figure 3. While this size distribution approximates a r^{-4} power law for radii between 0.1 and 20 μm,[44,45] there are some fluctuations about a slope of -4 because of the bimodal nature of the distribution.[46] The major change from the earlier version of the rural model[8] is that the number density of the very small ($r < 0.5\ \mu$m) particles is more accurately represented.

To allow for the dependence of the humidity effects on the size of the dry aerosol, the growth of the aerosol was computed separately for the accumulation and coarse particle components using Hänel's model No. 6 water uptake data. In accounting for the aerosol growth in Eq. (2), changes in the width of the size distribution were assumed negligible so only the mode radius, r_i, was modified by humidity changes. The effective refractive indexes for the two size components were then computed from Eq. (6) as a function of relative humidity. The cumulative number density and the volume distribution are shown in Figures 4 and 5 respectively for several different relative humidities. The refractive index as a function of wavelength and relative humidity is given in Table 4.

2.4 Urban Aerosol Model

In urban areas the air with a rural aerosol background is primarily modified by the addition of aerosols from combustion products and industrial sources. The urban aerosol model therefore was taken to be a mixture of the rural aerosol with carbonaceous aerosols. The sootlike aerosols are assumed to have the same size distribution as both components of the rural model. The proportions of the sootlike

aerosols and the rural type of aerosol mixture are assumed to be 20 percent and 80 percent respectively. The refractive index of the sootlike aerosols was based on the soot data in Twitty and Weinman's[47] survey of the refractive index of carbonaceous materials. As with the rural model, a composite urban aerosol refractive index was determined at each wavelength. These values are given in Table 3.

The change in aerosol was based on Hänel's[28] urban aerosol data (his Model 5) and is given in Table 2. The resulting refractive indexes as a function of relative humidity are given in Table 5.

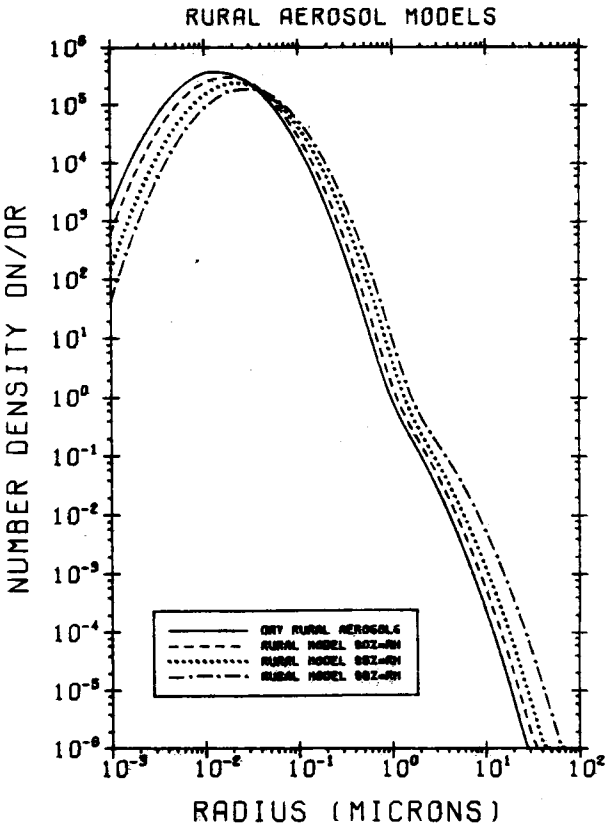

Figure 3. Aerosol Number Distribution (cm^{-3} μm^{-1}) for the Rural Model at Different Relative Humidities With Total Particle Concentrations Fixed at 15,000 cm^{-3}

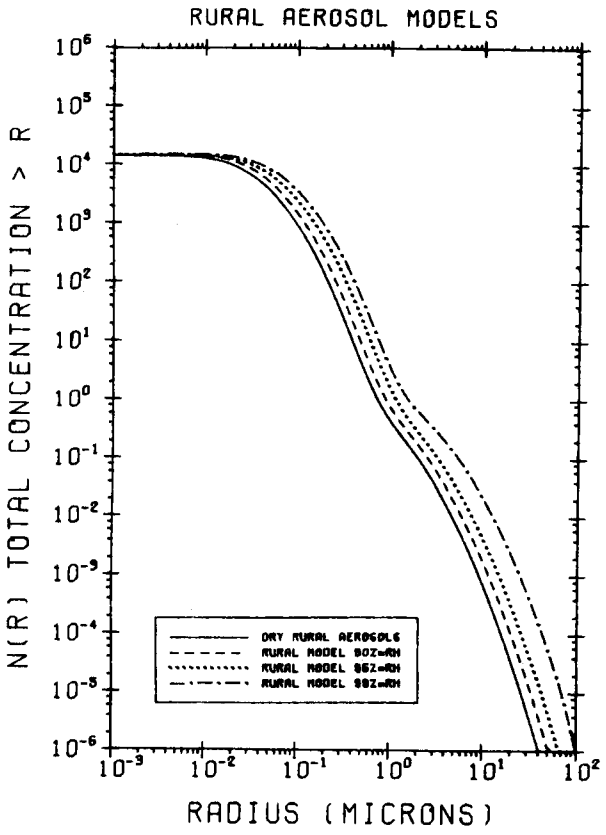

Figure 4. Cumulative Number Density (cm^{-3}) for the Rural Aerosol Model at Different Relative Humidities With Total Particle Concentration Fixed at 15,000 cm^{-3}

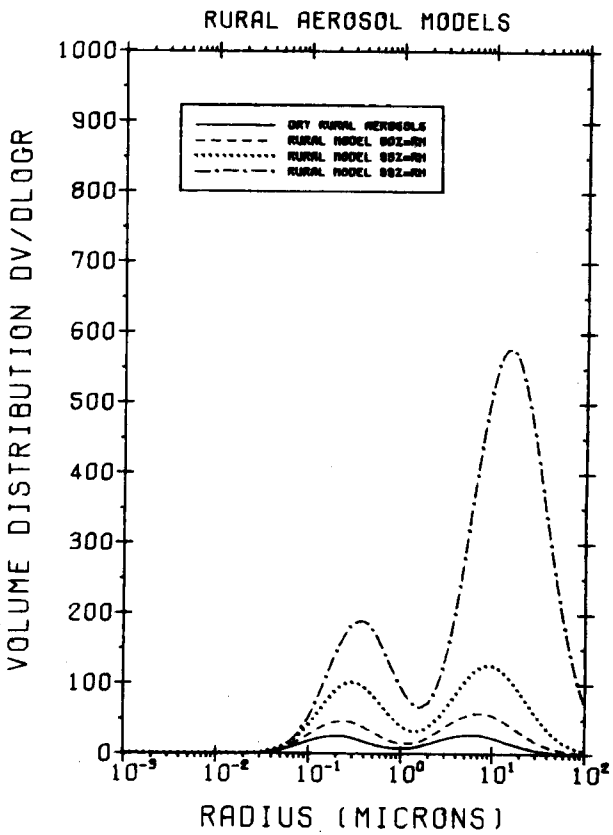

Figure 5. Volume Distribution ($\mu m^3/cm^3$) for the Rural Aerosol Model at Different Relative Humidities With the Total Particle Concentration Fixed at 15,000 cm^{-3}