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Relative — Humidity Parameterization of the Navy Aerosol Model (NAM)

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RELATIVE-HUMIDITY PARAMETERIZATION OF THE NAVY AEROSOL MODEL (NAM)

INTRODUCTION

The validity of the relative-humidity parameterization of the Navy aerosol model (NAM) [1] is evaluated and improved in this report for several reasons:

- The three lognormal size distributions corresponding to the three components of NAM consist in part of particles with sizes smaller than $0.1 \mu\text{m}$. This may be a source of error, because the existing humidity parameterization, based on a relationship given in Ref 2, only gives accurate results for particles greater than $0.1 \mu\text{m}$. Of the three components of NAM, the *continental or gas-to-particle component* is most susceptible to this error, since most of its particles are smaller than $0.1 \mu\text{m}$.
- The accuracy of the humidity relationship is limited to relative humidities (RHs) less than about 98% [2]. It would be desirable to extend the validity of NAM beyond 98%. While the RH range between 98% and 100% is narrow, and thus infrequently found in the maritime atmosphere, it is nevertheless important, because of the large changes in particle size at those humidities.
- A further improvement is to provide a separate humidity relationship for each component for which a different particle type is considered; presently Fitzgerald's relationship [2] for clean maritime aerosols (sea salt) is used for all three components.

Fitzgerald's relationship [2] gives the dependence of particle size on the dry size of the particle and on RH. It is an approximate form of the exact equation [3-5] for the equilibrium size of solution droplets. In arriving at his relationship, Fitzgerald assumes that the curvature effect (Kelvin effect) of the particles is negligible. This results in a numerically convenient formula but also causes the relationship to lose accuracy outside of the particle size and humidity ranges previously mentioned. Other approximate solutions to the exact growth equation have been proposed. Included are those found in Refs 4, 5, 6, 7, and 8. Those solutions are also unable to accurately predict the dependence of particle size on humidity for small particles and for high RH. Reference 5 gives predictions near $\text{RH} = 100\%$; however, none gives an analytical expression which is valid for all the particle sizes found in the NAM size distributions, and for the entire interval from 0% to 100%.

The use of the exact growth equations for a purpose such as NAM is also undesirable because the equations are implicit in particle size so they cannot be solved directly for particle size as a function of RH. The equations can be inverted to obtain particle size with an error minimization technique [9], or tables of particle-size values can be prepared. Neither method is useful for NAM where a simple explicit relationship which uses a minimum of computer time is preferred.

An improved approximation of the exact growth equations is described here. By including the influence of the Kelvin effect, this approximation avoids the shortcomings of the previous approximations. It gives acceptable accuracy in predicting particle size as a function of RH for dry particles ranging from 0.01 to $10 \mu\text{m}$, and for an RH range including all humidities up to 100%. The particles are

assumed to be chemically homogeneous, the hysteresis effect is not included, and the results are given for increasing humidity.

The new approximation is applied to the lognormal size distributions in NAM to discover their behavior as a function of RH, especially for the high values of RH and for the NAM component with the large fraction of particles less than $0.1 \mu\text{m}$. This behavior is used to parameterize the geometric mean and standard deviation of the lognormal distributions in terms of RH. The RH dependence of the aerosol optics (volume extinction coefficient) is estimated for the lognormal size distributions of NAM in which the full humidity parameterization as well as simplified versions which neglect the Kelvin effect are used. Limits are given for the applicability of the humidity parameterization for sea-salt, urban, and rural aerosols; and conclusions are drawn as to the validity of Fitzgerald's relationship [2].

LOGNORMAL SIZE DISTRIBUTIONS

The form of the lognormal distribution function used by Gathman [1] for NAM is

$$\frac{dN}{dr} = \sum_{i=1}^3 \frac{A_i}{f} \exp \left\{ -C \left[\ln r - \ln (f r_{0i}) \right]^2 \right\}, \quad (1)$$

where dN/dr is the number of particles per cm^3 per micrometer; i gives the number of the component in NAM with the size of the number in the same sequence as the size of the geometric means; r is particle radius in micrometers; r_{0i} is the mode radius; $C = 1/(2 \times s^2)$ where s is the standard deviation of the size distributions; A_i are parameters fitted to the meteorological conditions and particle concentrations of the NAM components [1]; and f is the RH effect given by the ratio of the particle radius at the ambient RH to the particle radius at the standard RH of 80%.

$$f = \left[\frac{2 - S}{6(1 - S)} \right]^{1/3}, \quad (2)$$

where S is the saturation ratio.

Rather than use Eq. (1) in this study, the normalized form of the lognormal distribution function [10] is used. It is given by

$$\frac{dN}{d \log r} = \sum_{i=1}^3 \frac{N_i}{(2\pi)^{1/2} \log b_g} \exp \left\{ -\frac{[\log r - \log (f r_{gi})]^2}{2(\log b_g)^2} \right\}, \quad (3)$$

where N_i is the number of particles per cm^3 for each component, b_g is the geometric standard deviation, and r_{gi} are the geometric means of the size distributions. Equation (3) was used, because for $N_i = 1$ integration over all particle sizes gives $dN/d \log r = 1$ which makes the comparison of distributions calculated for various values of RH, b_g , and r_{gi} easier, and permits the comparison of the present results with similar work which is also usually described in terms of b_g and r_g , e.g., [9].

Transformation equations [11] can be used to equate Eqs. (1) and (3). When this is done, $b_g/s = 2.87$, $r_{gi}/r_{0i} = 1.65$, $N_i/A_i = 1.38 \times r_{gi}$; and $b_g = 2.03$, $r_{g1} = 0.05 \mu\text{m}$, $r_{g2} = 0.4 \mu\text{m}$, and $r_{g3} = 3.3 \mu\text{m}$.

AEROSOL GROWTH EQUATION

The ratio of the water vapor pressure p'_r over the surface of an aerosol consisting of a solution droplet to the vapor pressure p_∞ over a plane water surface is given by the usual expression [4]

$$\frac{p'_r}{p_\infty} = \exp \left\{ \frac{2\sigma'}{r \rho' R_v T} \right\} \left[1 + \frac{l M_w \rho_d r_d^3}{M_s (r^3 \rho' - r_d^3 \rho_d)} \right]^{-1}, \quad (4)$$

where

- σ' is the surface tension of solution droplet;
- ρ' is the density of solution droplet;
- ρ_d is the density of dry hygroscopic particle;
- $R_v = 4.615 \times 10^6 \text{ erg g}^{-1} \text{ K}^{-1}$ is the specific gas constant of water vapor;
- T is absolute temperature;
- r is the radius of solution droplet;
- r_d is the radius of dry hygroscopic particle;
- i is the van't Hoff factor;
- M_w is the molecular weight of water; and
- M_d is the molecular weight of dry particle.

Under the assumption that the volumes of the water and the dry particle are additive in the solution droplet, the mass m_w of water in the solution droplet is given by

$$m_w = \frac{4\pi}{3} (r^3 \rho' - r_d^3 \rho_d) = \frac{4\pi}{3} (r^3 - r_d^3), \quad (5)$$

so that

$$\frac{m_d}{m_w} = \frac{r_d^3 \rho_d}{(r^3 - r_d^3) \rho_w}. \quad (6)$$

Using Hänel's [5] expression for σ' :

$$\sigma' = \gamma_\sigma [\sigma_w(T_0) + a(T_0 - T) + b m_d/m_w], \quad (7)$$

where $w(T_0) = 75.6 \text{ dynes/cm}$ is the surface tension of pure water at the temperature $T_0 = 273.16 \text{ K}$, $a = 0.153 \text{ dynes/cm}$, and γ_σ the effect on σ' due to surface effects is assumed to equal 1.00. The assumptions are made that $\rho' = \rho_w$ in the exponential term in Eq. (4); that the solution droplet is in equilibrium with the saturation ratio S of the atmosphere so that $S = p_r/p$; that the linear mass increase coefficient $\mu = i M_w/M_d$ tabulated for the sea-salt, urban, and rural particles by [5], and for the ammonium sulphate particles by [12] is independent of the r_d ; and that the dry particles are chemically homogeneous.

The preceding equations are combined to result in a form of the growth equation

$$S = \frac{\exp[(2\sigma')/(r \rho_w R_v T)]}{1 + \bar{\mu} m_d/m_w}, \quad (8)$$

which cannot be solved directly for r as expected, and which is used in the subsequent numerical calculations.

APPROXIMATE GROWTH EQUATION

Values of S were calculated by introducing a large number of closely spaced values of r , one value of r_d , constants for the particular aerosol type, and values of $\bar{\mu}$ into Eq. (8) at $T = 298 \text{ K}$. Since $\bar{\mu}$ is itself a function of S , an iterative procedure was used to choose the proper values of $\bar{\mu}$ in applying Eq. (8). Figure 1 shows the results of some of those calculations for ammonium sulphate solution droplets. Each solid line gives the calculated relationship between the water volume m_w and $-\log S$ for a particular ammonium sulphate r_d . By neglecting the Kelvin effect the relationship between m_w and $-\log S$ would appear in the form of straight lines in Fig. 1, which is approximately the case for large

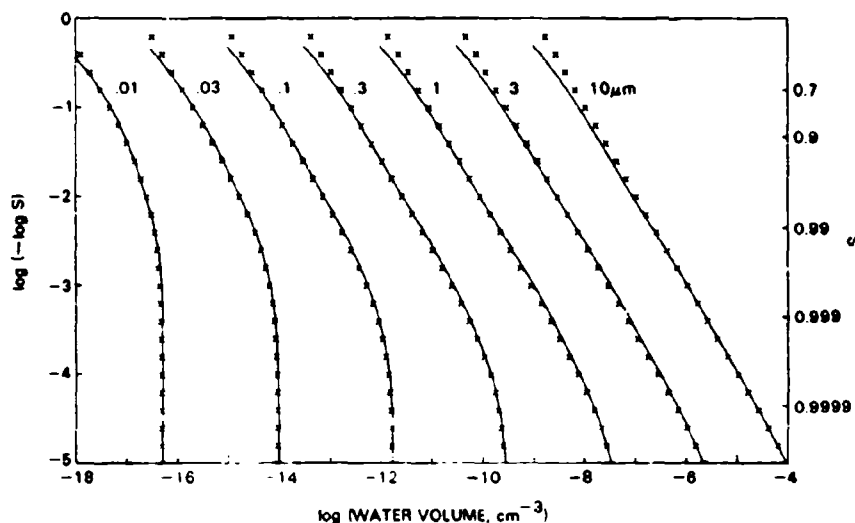


Fig. 1 — Volume of water in solution droplets containing ammonium sulphate with an initial dry-particle radius as given in equilibrium with the atmospheric water vapor saturation ratio S . The exact growth equation [solid curves, Eq. (8)] is compared with the approximate equation [x, Eq. (15)].

values of r as expected. The increasing nonlinearity of the curves for decreasing values of r_d demonstrates the importance of including the Kelvin effect for those dry particle sizes.

The analytical expression

$$y = \left[\frac{A}{x} - B \right] C \quad (9)$$

(where $x = m_w = 4\pi/3(r^3 - r_d^3)$ and $y = -\log S$) was fitted to the growth curves in Fig. 1.

Equation (9) is explicit in r so that it can be solved for r which gives

$$r = \left(\frac{A C D}{B C - \log S} + r_d^3 \right)^{1/3}, \quad (10)$$

where $D = 3/4\pi$.

Figure 2 shows that power laws relate A and C to r_d with good accuracy. Straight lines fitted through 13 geometrically equal values of r_d between 0.01 and 10 μm give

$$A = 1.490 \times 10^7 r_d^{4.510}, \quad (11)$$

and

$$C = 1.352 \times 10^{-7} r_d^{-1.428}. \quad (12)$$

The value of B is independent of r_d :

$$B = 2.30 \times 10^{-4} \quad (13)$$

and

$$D = 0.2387. \quad (14)$$

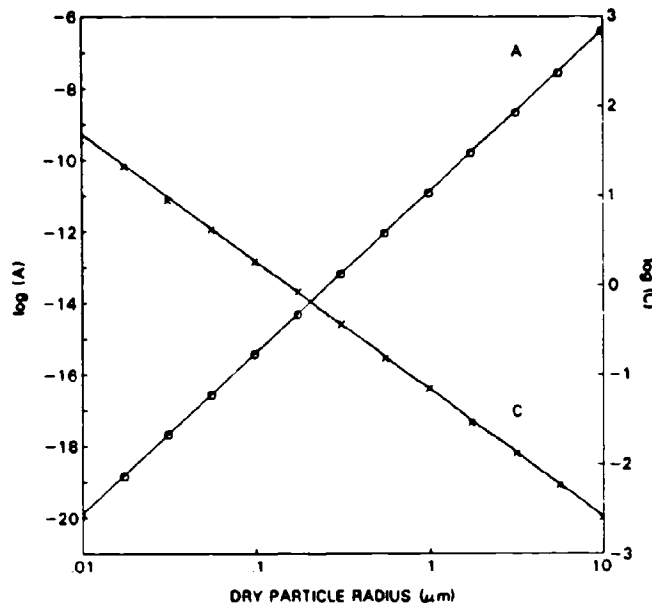


Fig. 2 — Relationships of A and C to the dry particle radius of ammonium sulphate particles [Eqs. (11), (12)]

Combining Eqs. (9)-(14) gives the final form of the approximate growth equation for ammonium sulphate particles:

$$r = \left(\frac{C1 r_d^{C2}}{C3 r_d^{C4} - \log S} + r_d^3 \right)^{1/3}, \quad (15)$$

where the constants $C1$, $C2$, $C3$, $C4$ are listed in Table 1. A similar fitting procedure was done for the sea-salt, rural, and urban aerosols; Table 1 also gives the appropriate constants.

Table 1 — Constants for the Growth Equation, Eq. (15); and for the Lognormal Size Distribution, Eqs. (19), (22), (27)

Aerosol Model	NAM Component	r_{gd} (cm)	b_{gd}	C1	C2	C3	C4	C5	C6
Sea Salt	2	2.460E-5	2.009	0.7674	3.079	2.572E-11	-1.424	1.411	10.23
Sea Salt	3	1.790E-4	2.009						
Urban	1	3.891E-6	1.997	0.3926	3.101	4.190E-11	-1.404	1.345	9.742
Rural	1	4.210E-6	2.002	0.2789	3.115	5.415E-11	-1.399	1.345	9.681
(NH ₄) ₂ SO ₄				0.4809	3.082	3.110E-11	-1.428		

The temperature dependence of Eq. (15) is found primarily in $C3$ due to the sensitivity of the Kelvin effect to temperature; $C3$ can be temperature corrected with the expression $C3(T) = C3[1 + 0.004(298 - T)]$.

Figure 1 illustrates the behavior of Eq. (15) for ammonium sulphate particles by values of m_w and $-\log S(x)$ calculated for each r_d curve. There is agreement between the exact [Eq. (8)] and the approximate [Eq. (15)] growth curves. In this case as well as for the other aerosol types, the largest differences occur for small values of S which fall below the deliquescent point of the particles. In this range rapid changes occur in the value of $\bar{\mu}$. The mean relative error in %, $[100 \times ABS(r(\text{approx.}) - r(\text{exact})) / r(\text{exact})]$, between the exact and approximate values of r for $S \geq 0.75$ and for each r_d

curve was calculated for all aerosol types and is given in Fig. 3. In all cases, the mean error is less than about 6% for the range of r_d between 0.01 and 10 μm . The maximum error of 13.5% was found for sea-salt particles with $r_d = 0.01 \mu\text{m}$ at $S = 0.75$.

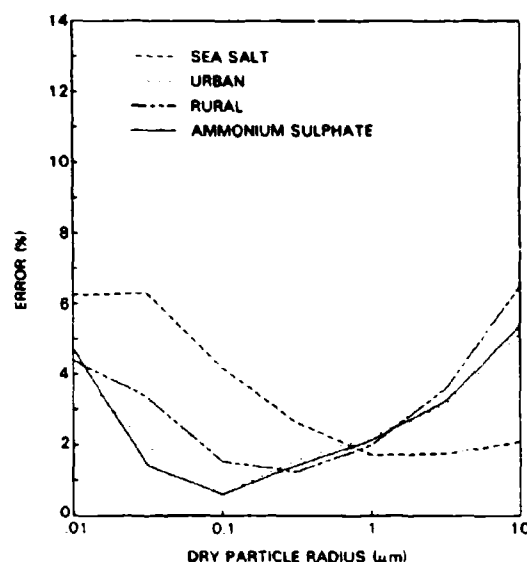


Fig. 3 — Mean error in the approximate growth equation as compared to the exact growth equation as a function of the dry particle radius and the different aerosol models

RELATIVE-HUMIDITY PARAMETERIZATION OF THE LOGNORMAL SIZE DISTRIBUTIONS OF NAM

Using the approximate growth formula [Eq. (15)], the behavior of the lognormal size distributions [(Eq. (3))] of NAM can be investigated as a function of S . Given that Eq. (3) is in terms of r_d , and the dry-particle geometric mean r_{gd} and standard deviation b_{gd} , and that $f=1$, the particle size distribution as a function of S and r is given by

$$\frac{dN}{d\log r_j} = \frac{dN}{d\log r_{dj}} \frac{d\log r_{dj}}{d\log r_j}, \quad (16)$$

where

$$d\log r_{dj} = \{\log(r_{dj+1}/r_{dj-1})\}/2 = \text{constant}, \quad (17)$$

$$d\log r_j = \{\log(r_{j+1}/r_{j-1})\}/2, \quad (18)$$

and j is the index for the particle radius.

Figure 4 shows the results of applying Eq. (16) to the mid-sized component of NAM consisting of sea salt with $r_{gd} = 0.246 \mu\text{m}$ and $b_{gd} = 2.009$ (these values are both smaller than the values of r_g and b_g given previously which were for $S = 0.8$). Several changes are evident in the size distributions in Fig. 4 as S increases: the maximum value of $dN/d\log r$ shifts to larger particle sizes and at the same time decreases somewhat, and the width of the distributions increases. The increase in width means that $d\log r$ is no longer a constant like $d\log(r_d)$ but changes with S . This clearly shows that the mean as well as the standard deviation of the distributions are a function of S .

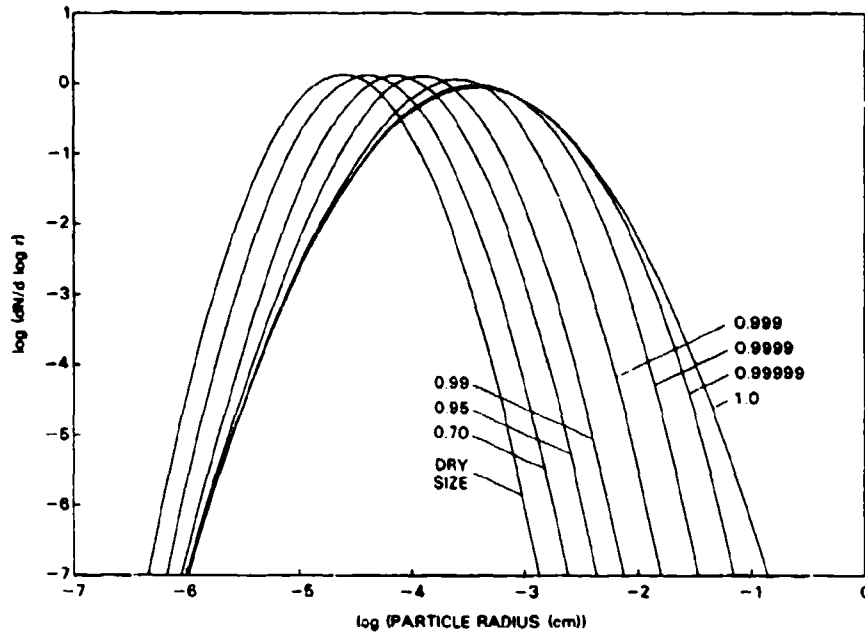


Fig. 4 — Dependence of the lognormal mid-sized sea-salt component of NAM on the given saturation ratios

A large number of calculations were required to obtain the curves in Fig. 4, because 100 values of r_d were needed for each decade in particle size to achieve acceptable accuracy. That makes the use of Eq. (16) less than desirable for an application such as NAM. For this reason, an attempt was made to describe the distributions in Fig. 4 with a more practical expression given by a form of the lognormal distribution

$$\frac{dN}{d \log r} = \sum_{i=1}^3 \frac{N_i}{(2\pi)^{1/2} \log b_g(S)} \exp \left\{ - \frac{[\log r - \log r_g(S)]^2}{2[\log b_g(S)]^2} \right\}, \quad (19)$$

where both r_g and b_g are functions of S . This equation should be used in NAM instead of Eqs. (1) or (3) when particle-size information is desired at all humidities including 100%.

It is also apparent in Fig. 4 that some distortion results in the originally lognormal distribution for dry particles as the value of S increases. This comes about because the stronger influence of the Kelvin effect for the small dry particles prevents them from growing as large as the larger dry particles. Thus before Eq. (19) can be considered for inclusion in NAM, it is necessary to obtain an estimate of how badly the original lognormal distribution is distorted. This can be done by calculating $r_g(S)$ and $b_g(S)$ for the actual distributions given by Eq. (16) and comparing those distributions with Eq. (19). The expressions for $r_g(S)$ and $b_g(S)$ are

$$r_g(S) = 10^{\left[\frac{\sum \log r_j}{\sum \frac{dN}{d \log r_j} d \log r_j} \right]}, \quad (20)$$

and

$$b_g(S) = 10^{\left[\frac{\sum \frac{dN}{d \log r_j} d \log r_j (\log r_j - \log r_g(S))^2}{\sum \frac{dN}{d \log r_j} d \log r_j} \right]^{1/2}}. \quad (21)$$

Figure 5 shows the comparison of the actual distributions to the lognormal distributions which are in terms of $r_g(S)$ and $b_g(S)$. The distortion is relatively small for $S < .99$, it reaches a maximum near $S = .9999$, and is negligible for $S = 1.00$. Under maximum distortion, values of r predicted by Eq. (19) are accurate near maximum values of $dN/d\log r$, these values lose accuracy as $dN/d\log r$ decreases, and they are in error by about 30% for $dN/d\log r$ reduced three orders of magnitude from its peak value. This value of S for the maximum distortion only applies to the mid-sized sea-salt component. It shifts to a value of $S = 0.999$ for the component with the smallest particle sizes and to $S = 0.99999$ for the component with the largest particle sizes. In all cases the distortion is negligible at $S = 1.00$. While this distortion is undesirable, it occurs at values of S which will seldom be used due to their large sizes.

Equations (20) and (21) are also too computationally involved to be practical with NAM; hence, approximate forms were found. The approximate expression for $r_g(S)$ is simply given by Eq. (15) written in terms of the dry geometric mean radius r_{gd} :

$$r_g(S) = \left(\frac{C1 r_{gd}^2}{C3 r_{gd}^4 - \log S} + r_{gd}^3 \right)^{1/3} \quad (22)$$

Equation (22) predicts the value of $r_g(S)$ with an accuracy of better than 3.2% over the entire range of S and for all three aerosol components.

An approximate expression for Eq. (21) is not as straightforward, because of the complex behavior of $b_g(S)$ as a function of S as shown in Fig. 6. A reasonable fit to the solid lines given by Eq. (21) is the tangent function in the form

$$y = -[A + B \tan(Cx)] \quad (23)$$

where

$$x = \pi \left(\frac{b_g(S) - b_{gd}}{b_g(1.0) - b_{gd}(\text{dry})} - \frac{1}{2} \right) \quad (24)$$

$$y = \log(1 - S) \quad (25)$$

and

$$A = C5 \log r_{gd} + C6 \quad (26)$$

$B = 0.65$ and $C = 0.88$ are empirical constants, and $C5$ and $C6$, listed in Table 1, are constants which depend on the aerosol component.

Given that in all cases $b_g(1.0) - b_{gd} \approx 0.85$, Eqs. (23) to (26) are combined to give the desired approximate relationship for $b_g(S)$:

$$b_g(S) = b_{gd} + 0.300 \arctan \left[- \frac{C5 \log r_{gd} + C6 + \log(1 - S)}{0.650} \right] + 0.415 \quad (27)$$

Figure 6 shows the predictions of Eq. (27) for the sea-salt component which was assumed to apply to all three lognormal size distributions of NAM. The accuracy of Eq. (27) is better than 0.05 for sea salt as well as for the other two components which give similar results. Figure 6 also demonstrates that the change of the geometric standard deviation is limited to large values of RH. For the lognormal distribution with the smallest particle sizes the geometric standard deviation must be corrected with Eq. (27) for values of S greater than about 0.90, while for the other two distributions the correction occurs at higher values of S .

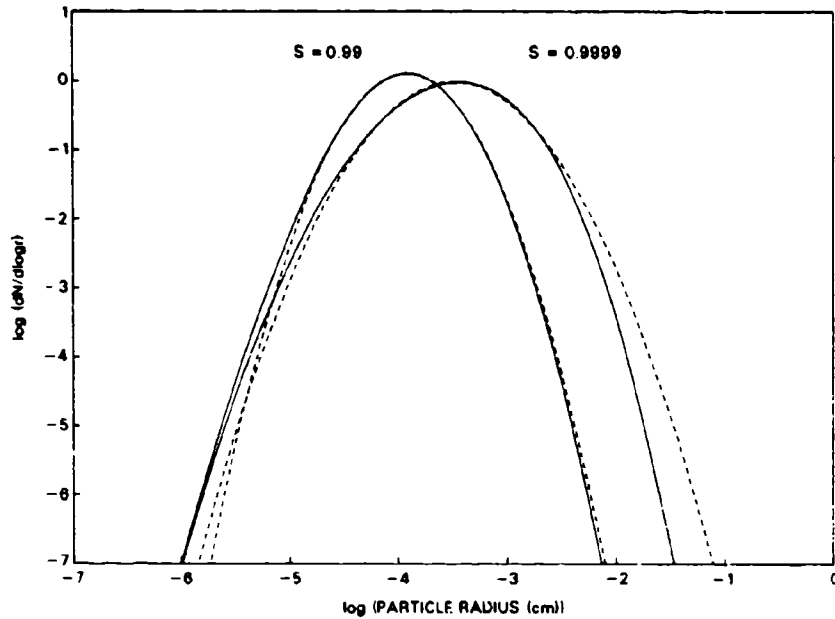


Fig. 5 — Distortion of the sea-salt size distributions at large values of S as shown by the actual distributions (solid lines) and the best fit lognormal distributions (dashed lines)

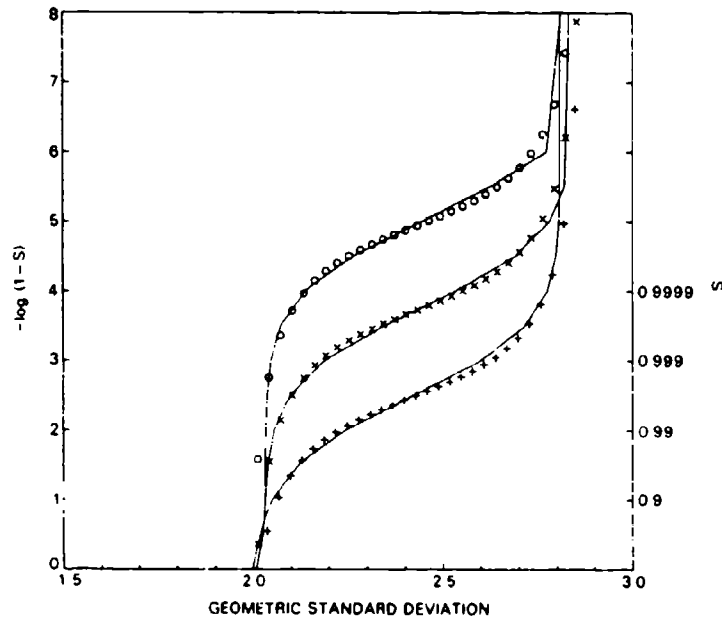


Fig. 6 — Change in the geometric standard deviation of the lognormal size distributions of NAM as a function of the saturation ratio S . All data correspond to sea-salt particles. Solid curves are from exact calculations, data points are from an approximation relationship [Eq. (27)]; and the three data sets correspond to the smallest (+), mid-sized (x), and largest (o) NAM components.

OPTICAL PROPERTIES COMPUTED FROM THE LOGNORMAL SIZE DISTRIBUTIONS OF NAM

Until now, this analysis has dealt only with the RH parameterization of the size distributions of NAM, with Eq. (19) giving the final form. Since the ultimate objective of NAM is to predict the optics of the aerosol particles, this section investigates the accuracy of the aerosol optics calculated for Eqs. (3) and (19). Equation (3) is the transformed version of Gathman's [1] equation [Eq. (1)] used in NAM. Equation (19) should be more accurate for calculating the optical effects than Eq. (3); however, it has not been made clear what the accuracy of the optics are using Eq. (3). This accuracy should be better than the limits imposed on the applicable range of RH [Eq. (2)] by Fitzgerald [2], because the optical properties are primarily determined by particles greater than $0.1 \mu\text{m}$ for which the Kelvin effect has less of an influence.

To determine the accuracy of Eqs. (3) and (19) for calculating the aerosol optics a complete Mie calculation is not done. Instead the cross section for extinction of the aerosol particles is assumed to be proportional to the particles' area. This assumption gives an estimate of the upper limit of the error in using those equations for the aerosol optics, and it permits much shorter and simpler calculations. The area of the particles described by the size distribution in Eq. (16) is calculated and is assigned to be exactly proportional to the extinction coefficient of the particles. Areas (extinction coefficients) calculated for the approximate expressions of the size distributions are then compared to the exact extinction coefficient.

The comparison of the Eq. (19) [including Eqs. (22) and (27)] to Eq. (16) as a function of RH and the NAM components gives the following results: the discrepancy between the extinction coefficients determined from those equations is small except where the distortion of the size distribution given by Eq. (19) is large. This occurs at very high values of RH as discussed before. In this region errors on the order of 15% are found. For even larger values of RH the errors decrease, and disappear altogether at RH = 100%.

Equation (19) is next simplified by removing the Kelvin effect [found in the term $C3 r_{gd}^{C4}$ of Eq. (22)], and by assuming that $b_p(S)$ is simply a constant equal to 2.03. The particles' area resulting from this modified form of Eq. (19) is again compared to the area of Eq. (16) with the following results: differences do not exceed about 10% for RH < 99% for the urban and rural aerosol (component 1), for RH < 99.9% for the sea-salt particles (component 2), and for RH < 99.99% for the sea-salt particles (component 3).

If the additional approximations

$$r_{gd}^{C2} \approx \text{constant} \times r_{gd}^3 \quad (28)$$

and

$$-\log S \approx 1 - S \quad (29)$$

are made to Eq. (22), then

$$\frac{r_g(S)}{r_{gd}} = \left(1 + \frac{\gamma}{1 - S} \right)^{1/3}, \quad (30)$$

which is equivalent to Eq. (7) in Fitzgerald's [2] report, and where γ is defined by Fitzgerald [2] as the air-mass characteristic. For the sake of comparison with his values of γ , the values of γ found here are 0.825, 0.965, 0.282, and 0.173 for sea-salt particles (components 2 and 3) and for urban and rural particles. These values agree reasonably well with Fitzgerald's [2] values. It is of interest to note that the value of γ differs for the two sea-salt components. This demonstrates that in the growth curve [Eq. (30)] the air-mass characteristic γ depends not only on the origin of the particles, but also on their size distribution.

For $RH = 80\%$ ($S = 0.80$) Eq. (30) becomes

$$r_g(0.80) = r_{gd} \left(1 + \frac{\gamma}{0.2} \right)^{1/3} \quad (31)$$

Dividing Eq. (30) by Eq. (31) eliminates the dependence on r_{gd} and gives

$$f = \frac{r_g(S)}{r_g(0.8)} = \left(\frac{C7 - S}{C8(1 - S)} \right)^{1/3} \quad (32)$$

which is the form used by Gathman [1] in NAM. Table 2 lists the values of $C7$, $C8$, and the range of validity of Eq. (32) for the various NAM components. The values of $C7 = 2$ and $C8 = 6$ used by Gathman [1] for all NAM components are close to the present values for the sea-salt particles in NAM component 3. The assumptions in Eqs. (28) and (29) add only a small error to Eq. (32). This means that extinction coefficients calculated with Eqs. (19) and (32), which give an expression equivalent to Eq. (3), have an accuracy of better than about 10% for the range of validity (upper limit) shown in Table 2. Thus under most situations found in the maritime atmosphere, Eq. (3) [with f given by Eq. (32)] can be used instead of Eq. (19). The constants in Table 2 must be used with Eq. (3), and the range of validity should not be exceeded, since large errors are found at greater values of RH .

Table 2 — Constants for Eq. (32), and the Validity of Eq. (3) for Calculating Extinction Coefficients.

Aerosol Model	NAM Component	C7	C8	Range of Validity
Sea Salt	2	1.83	5.13	$RH < 99.9\%$
Sea Salt	3	1.97	5.83	$RH < 99.99\%$
Urban	1	1.28	2.41	$RH < 99\%$
Rural	1	1.17	1.87	$RH < 99\%$

SUMMARY AND RECOMMENDATIONS

A new approximation formula for the equilibrium size of hygroscopic particles as a function of RH and particle type was developed. This formula includes the influence of the Kelvin effect which gives the formula a range of applicability much larger than previous approximation formulas. It applies to particles with radii extending from 0.01 to 10 μm , and for a RH range from 0% to 100%. The new formula was applied to the size distributions of NAM to discover the effect of having neglected the Kelvin effect previously, and the behavior of the distributions at high values of RH . A new version of the NAM size distributions [Eq. (19)] was developed where the geometric mean and standard deviation were both parameterized as a function of RH . It is estimated that the error of calculating aerosol extinction coefficients with Eq. (19) does not exceed about 20% over the entire RH range from near 0% to 100%, and is significantly less for most values of RH , when all sources of error are considered. Sources of error are found in the approximation formula for particle growth primarily due to the non-linear behavior of the mass increase coefficient, in the approximations used for the geometric mean and standard deviation of the size distributions, and in the distortion of the lognormal distributions at very high values of RH . This accuracy is sufficient for NAM, since much greater uncertainties are caused by the mass increase coefficients given by Hänel [5] which reflect average conditions and thus do not include the large fluctuations in the coefficients which could occur as a function of time.

It was demonstrated that the humidity parameterization in Eq. (19) reduced to the one presented by Fitzgerald [2], [Eq. (30)] and used by Gathman [1], [Eq. (32)]. The reduction was accomplished by applying a series of approximations which included neglecting the Kelvin effect and assuming that the

geometric standard deviation was independent of RH. The error in using Gathman and Fitzgerald's humidity parameterized size distribution [Eqs. (1) and (2)] for calculating extinction coefficients for NAM also does not exceed 20%, except for values of RH which are larger than those listed in Table 2.

Specific recommendations for changes to NAM in LOWTRAN are:

- For both sea-salt components no changes are required. The difference in the values of the coefficients ($C7$ and $C8$) in the growth equation [Eq. (32)] determined here and those used by Gathman [1] in Eq. (2) are small and cause negligible error in aerosol optics calculations.
- For the continental or gas-to-particle component of NAM, $C7$ and $C8$ in Table 2 should be used with Eq. (2) [see Eq. (32)]. Optical coefficients should be calculated for the urban and rural aerosols, and criteria established for their use within NAM.
- The routine use of NAM in LOWTRAN is limited to $RH < 99\%$, because over this RH range Gathman's (1983) formulation [Eq. (1)] can be retained in NAM, and the humidity parameterization gives valid results for all components.
- The description of NAM in LOWTRAN should reference this report to indicate that it gives approximation formulas which extend the RH dependence of the sea-salt, urban, and rural particles to $RH = 100\%$. This extension is appropriate for special studies, such as for example, particle behavior under prefog or precloud conditions.

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