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THE EFFECTS OF RELATIVE HUMIDITY ON ATMOSPHERIC AEROSOL EXTINCTION IN THE 3-5μm AND 8-15μm SPECTRAL REGIONS

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W. G. Tam [redacted] C. Boily

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RESUME

L'étude du changement du coefficient d'extinction des aérosols infrarouges en fonction de l'humidité relative est l'objet de ce rapport. On se base sur un modèle à deux types de distribution des particules: maritime et continental. Le facteur du coefficient d'extinction des aérorols s'accroît de plus de 10 dans les bandes spectrales de 3-5 et 8-15 µm lorsque l'humidité relative passe de 0.5 à 0.99. Aux humidités relatives très élevées, une région d'extinction plus faible est observée autour de 10-11 µm. (NC)

ABSTRACT

The variation of the infrared aerosol extinction coefficient as a function of relative humidity is studied. This is based on a two-component particle distribution model. The aerosol extinction coefficient increases by a factor of more than 10 throughout the 3-5 and 8-15 ~~µm~~ micrometers spectral regions as the relative humidity increases from 0.5 to 0.99. For high relative humidities, a region of lower extinction around 10-11 ~~µm~~ micrometers is noticed. (U)

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1.0 INTRODUCTION

The attenuation of an electromagnetic wave propagating in the atmosphere arises from molecular and aerosol extinctions. In the 3-5 μm and 8-15 μm 'window' regions where no major molecular resonance absorption is present, aerosol extinction is of primary importance.

With a change of relative humidity in the atmosphere, condensation or evaporation of water takes place on the aerosol particles and these, at the same time, change the particle optical parameters which determine the aerosol extinction. When the relative humidity increases, the size of an aerosol particle increases through the accretion of water. A particle can triple or quadruple its dry-state size at relative humidities above 95%. Together with the growth in size, the complex refractive index of the particle varies. Using a two-component distribution model, Barnhardt and Streete [1] have calculated the aerosol scattering coefficient in the infrared as a function of relative humidity. This work has been extended by Hodges [2] to include the absorption of the aerosols. However, a basic assumption made in both works is that aerosol particles grow at a rate dependent only on the relative humidity and not on the chemical composition of the particles in the original dry state. A recent comprehensive study has been carried out by Hänel [3] providing empirical data on the change of aerosol optical parameters as functions of relative humidity through the concept of the mass increase coefficient. By means of these empirical results, a more realistic assessment of the effects of relative humidity on aerosol extinction can now be made.

In practice, the aerosol extinction coefficients over the 3-5 μm and 8-15 μm spectral regions are of particular importance to the operation of broad-band infrared systems. In this report, we present the aerosol extinction coefficients as functions of relative

humidity in the above two window regions obtained, using the two-component particle number distribution model of [1] and the empirical data of the mass increase coefficients of [3]. This enables us to take into account the different behavior of the maritime and continental aerosols with the change of the relative humidity.

The work was performed at DREV between January and November 1977 under PCN 33A11 "Studies of Aerosols".

2.0 AEROSOL EXTINCTION AND RELATIVE HUMIDITY

Let $n(f, r)$ be the number of aerosol particles per unit volume with radius between r and r + dr at a given relative humidity f. The corresponding aerosol extinction coefficient $\beta(f, \lambda)$ for electromagnetic radiation of wavelength λ is given by

$$\beta(f, \lambda) = \pi \int_0^\infty n(f, r)\, Q_{ext}\left[\alpha, \eta(f, \lambda)\right] r^2 dr \qquad (1)$$

where $\alpha = 2\pi r/\lambda$ is the size parameter; $\eta(f, \lambda)$, the complex refractive index and Q_{ext}, the efficiency factor of extinction.

We wish to consider the problem by starting from the situation with f = 0*. The dry-state condensation nuclei distribution is $n(f = 0, r)$. To find the extinction coefficient at any other relative humidity $f > 0$, it is necessary to relate $n(f, r)$ to $n(0, r)$ and $\eta(f, \lambda)$ to $\eta(0, \lambda)$. To concentrate our attention on the effects of the change of relative humidity, we assume that as f increases $n(f, r)$ changes only by virtue of the condensation of water vapor. This is to say that all the particles retain their individuality without coagulation and furthermore, no new particles are produced in the meantime.

* We have formulated our problem in such a manner as to exclude the consideration of the hysteresis effect. $\beta(f, \lambda)$ is, in general, not a single-valued function of f. A detailed discussion can be found in [3]

We assume, following Barnhart and Street [1], that the particle number density n(f, r) consists of the continental and maritime components with a variable mix ratio so that

$$n(f, r) = \upsilon n_C(f, r) + (1 - \upsilon)\, n_M(f, r) \tag{2}$$

In eq. (2), $n_C(f. r)$ and $n_M(f, r)$ are the particle-number densities of the continental and maritime components respectively. The quantity $\upsilon (0<\upsilon<1)$ is the mix ratio.

In references [1] and [2], it was assumed that at a given relative humidity, the rates of growth of all the aerosol particles were equal. However, because of their different chemical compositions, the growth rate of a deliquescent sea salt is significantly different from that of the condensation nuclei of continental origin. In fact, it has been found experimentally [3] that $\mu(f)$, the linear mass-increase coefficient of an aerosol sample, given by

$$\mu(f) = \frac{m_W}{m_o} \quad \frac{1 - f}{f} \tag{3}$$

where m_o is the mass of the dry-state aerosol particles and m_W, the mass of water uptake of the sample, can be very different for different types of aerosols. For an aerosol particle of radius r_o in the dry state, Hänel [3] has obtained the following relationship giving its radius r at relative humidity f:

$$r = r_o \left[\left(1 + \frac{\rho_o}{\rho_w}\, \mu(f)\, \frac{f}{1 - f} \right)^{1/3} - \frac{1.056 \times 10^{-7}}{r_o} \quad \frac{f}{1 - f} \right.$$

$$\left. \left(1 + \frac{\rho_o}{\rho_W}\, \mu(f)\, \frac{f}{1 - f} \right)^{-1/3} \right] \tag{4}$$

The symbols ρ_o and ρ_W in eq. (4) denote the density of the dry-state aerosol and water respectively. The second term on the right-hand side

accounts for the correction due to the finite radius of the particle. In many cases, the correction term can be neglected. As for the refractive index $\eta(f, \lambda)$, it is related to the dry-state refractive index of the aerosol $\eta(f = 0, \lambda)$ and $\eta_W(\lambda)$ of water by

$$\eta(f, \lambda) = \eta_W(\lambda) + \left[\eta(f = 0, \lambda) - \eta_W(\lambda)\right] \left(1 + \frac{\rho_o}{\rho_W}\ \mu(f)\ \frac{f}{1 - f}\right)^{-1} \qquad (5)$$

3.0 CALCULATION OF THE AEROSOL EXTINCTION COEFFICIENTS

As mentioned in the previous section, the aerosol number density $n(f, r)$ consists of two components. The continental and maritime components are chosen such that at $f = 0.7$ they are the same as those used by Barnhardt and Streete in [1]. By means of numerical fitting we obtain

$$n_C(f = 0.7, r) = 0.258\ r^{-3.98} \qquad (6)$$

and

$$n_M(f = 0.7, r) = 0.369\ r^{-1}\ e^{-0.748\ r} \qquad (7)$$

Once the linear mass-increase coefficients $\mu(f)$ are given for the continental and maritime components, eqs. (4), (6) and (7) enable us to calculate $n(f, r)$ for other values of the relative humidity f.

For the linear mass-increase coefficients $\mu(f)$, we have chosen the experimental data of the sea-spray aerosol [4] i.e. (Model 2 of [3]) for the maritime component and that of alpine aerosol, (Model 6 of [3]) for the continental components. The values of $m(f)$ we used are in Table I.

TABLE I

Linear Mass-Increase Coefficients

f	μ(f)	
	Maritime	Continental
0.5	0.108	0.032
0.6	0.136	0.04
0.7	0.216	0.0505
0.8	0.782	0.14
0.9	0.617	0.15
0.95	0.548	0.105
0.99	0.468	0.117

At f = 0.99, the uptake of water is 46.33 times the mass of the dry-state aerosol for the maritime component, while it is 11.58 times for the continental component. Hence, the maritime aerosol particles grow much more rapidly than the continental ones.

Barnhardt and Streete [1] have shown that to calculate β in the infrared region it is sufficient to consider aerosol particles with radii in the range from 0.1 μm to 20 μm. In this size range, the correction term due to the finite radius of the particles in eq. (4) is always less than 1% for both the maritime and continental components. Thus, this term will be neglected. To make our results more readily comparable with those of [1] and [2], we take the lower limit of the integral of (1) to be 0.1 μm for all values of f, and the upper limit equal to 20 μm when measured at f = 0.7.

Before evaluating the efficiency factor for extinction $Q_{ext}\ [\alpha, \eta(f, \lambda)]$, it is necessary to have the complex refractive index $\eta(f, \lambda)$. The quantities η_W, $\eta(f = 0, \lambda)$ entering eq. (5) are summarized in Table II. In this table, the real part η_o' of the index

of refraction of the maritime aerosol component in the dry state is taken to be that of NACℓ as given in [5]. The imaginary part η_o'' is obtained from the absorption measurement of sea-salt carried out by Volz [6]. The complex refractive index for the continental aerosol component in the dry state is that used in [7]. The index of refraction of water is taken from the results compiled by Hale and Querry [8]. Figures 1a to 1d show the refractive indices of the continental and maritime aerosols for various values of f as obtained from eq. (5), and the corresponding dry-state values.

The dry-state densities are taken to be ρ_o(maritime) = 2.25 gm/c.c. and ρ_o(continental) = 1.8 gm/c.c.

TABLE II

λ	η_o'	η_o''	η_o'	η_o''	η_w'	η_w''
	Maritime		Continental		Water	
3	1.5242	.0035	1.3420	.0190	1.3710	.2720
3.5	1.5230	.0014	1.3990	.0068	1.4000	.0094
4	1.5217	.0029	1.3965	.0071	1.3510	.0046
4.5	1.5204	.0033	1.4000	.0133	1.3320	.0134
5	1.5188	.0028	1.3870	.0161	1.3250	.0124
8	1.5064	.0179	1.1387	.0691	1.2910	.0343
8.5	1.5038	.0307	1.3000	.1775	1.2780	.0367
9	1.5009	.0299	2.3020	.3010	1.2620	.0399
9.5	1.4979	.0202	1.8840	.1606	1.2430	.0444
10	1.4947	.0179	1.7990	.0696	1.2180	.0508
10.5	1.4914	.0167	1.7305	.0826	1.1850	.0662
11	1.4878	.0166	1.6900	.0665	1.1530	.0968
11.5	1.4840	.0172	1.6612	.0670	1.1260	.1420
12	1.4800	.0180	1.6325	.0675	1.1110	.1990
12.5	1.4758	.0190	1.6037	.0680	1.1230	.2590
13	1.4696	.0206	1.5750	.0685	1.1460	.3050
13.5	1.4668	.0228	1.5406	.0772	1.1770	.3430
14	1.4619	.0273	1.5061	.0860	1.2100	.3700
14.5	1.4568	.0311	1.4717	.0947	1.2410	.3880
15	1.4514	.0387	1.4650	.1700	1.2700	.4020

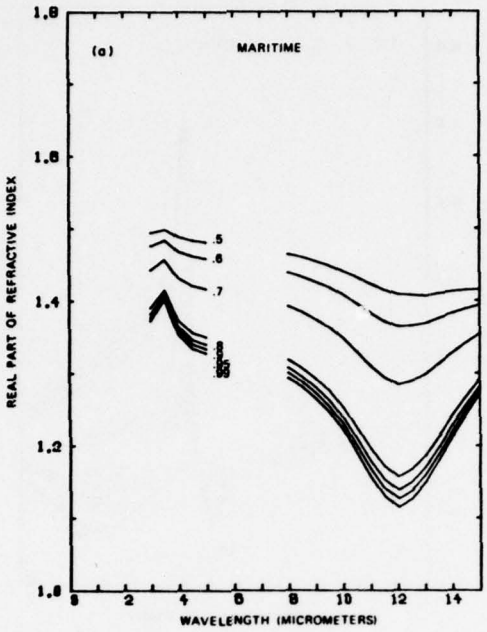

FIGURE 1(a) - Refractive index (real part) as a function of wavelength for maritime aerosols. The number accompanying each curve is the relative humidity.

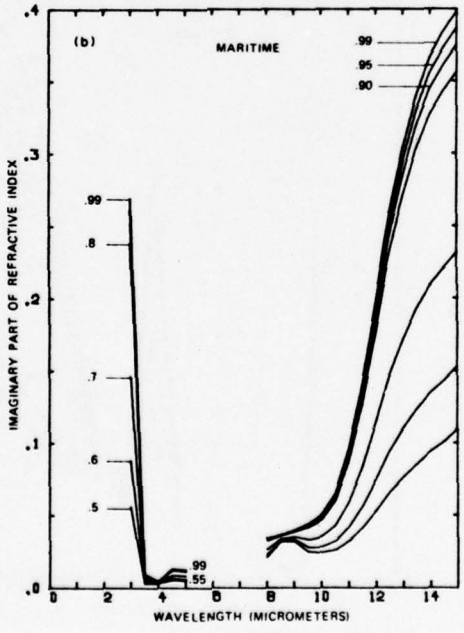

FIGURE 1(b) - Refractive index (imaginary part) as a function of wavelength for maritime aerosols. The number accompanying each curve is the relative humidity.

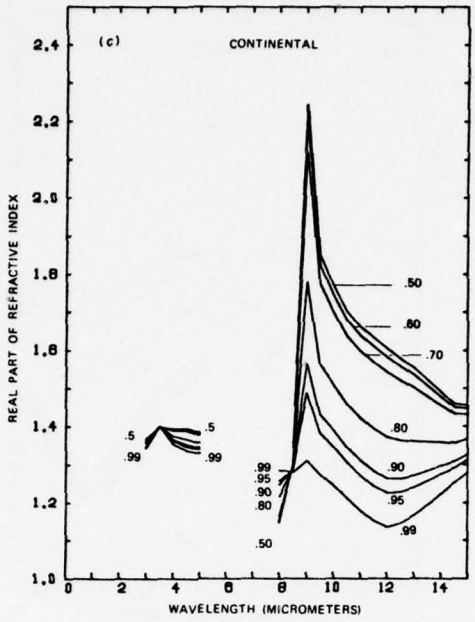

FIGURE 1(c) - Refractive index (real part) as a function of wavelength for continental aerosols. The number accompanying each curve is the relative humidity.

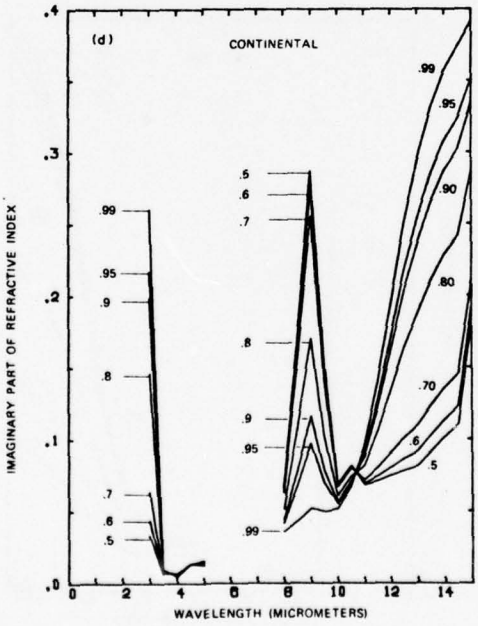

FIGURE 1(d) - Refractive index (imaginary part) as a function of wavelength for continental aerosols. The number accompanying each curve is the relative humidity.

Let $\mu_C(f)$ and $\mu_M(f)$ be the linear mass-increase coefficient of the continental and maritime components respectively and $F_C(f)$, $F_M(f)$ be the corresponding growth factors defined by

$$r = r_o F_C(f) \tag{8}$$

$$r = r_o F_M(f) \tag{9}$$

Equation (8) describes the growth of an aerosol particle of the continental component with a dry-state radius r_o and eq. (9) is the corresponding relation for an aerosol particle of the maritime component. Thus,

$$F_C(f) = \left(1 + \frac{\rho_o}{\rho_W} \mu_C(f) \frac{f}{1 - f} \right)^{1/3} \tag{10}$$

and

$$F_M(f) = \left(1 + \frac{\rho_o}{\rho_W} \mu_M(f) \frac{f}{1 - f} \right)^{1/3} \tag{11}$$

The functions $F_C(f)$, $F_M(f)$ are shown in Fig. 2 together with the growth factor used in [1]. From the assumption that no aerosol particles are created or removed, the number density $n(f, r)$ given in eq. (2) can be related to $n_C(f = 0.7, r)$ and $n_M(f = 0.7, r)$ as follows:

$$n(f, r) = \upsilon\, n_C\left(f = 0.7, \frac{F(f - 0.7)}{F(f)} r\right) + (1 - \upsilon)\, n_M\left(f = 0.7, \frac{F(f = 0.7)}{F(f)}\right) \tag{12}$$

The calculations of the efficiency factors of extinction can be carried out using the Mie theory [9]. For this purpose, we have used the program of Dave [10]. For small values of the size parameter α, Rayleigh's approximation suffices. We have used this approximation for $r < 0.5$ μm. The integration in eq. (1) is then performed by means of Simpson's rule.

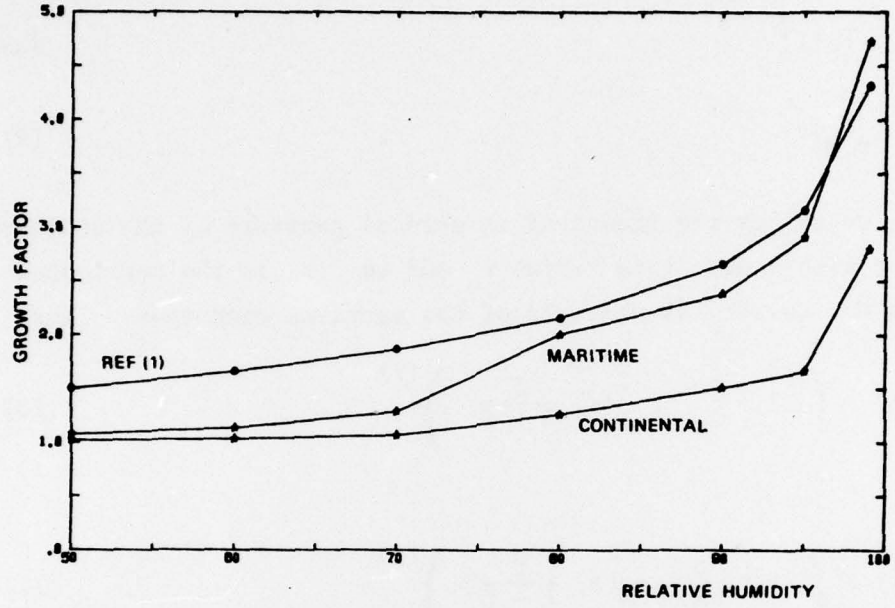

FIGURE 2 - Growth factor for maritime and continental aerosols and that used by reference [1] as a function of relative humidity.

4.0 RESULTS AND DISCUSSIONS

In Figs. 3 and 4, the results for the aerosol extinction coefficient β for the maritime and continental components are shown for various values of relative humidity f. For the maritime component, β is a monotonic decreasing function of λ in the wavelength regions considered for f = 0.5 and 0.6. When f increases beyond 0.7, a broad minimum occurs around 11 μm.

For the continental component, because of the large refractive index of the dry-state aerosol around 9 μm, the extinction coefficient shows a marked peak in this spectral region at low relative humidity. At higher relative humidity, this feature is washed out with the accretion of water and a broad minimum again appears around 10 μm.

In Figs. 5-11, the aerosol extinction coefficient for the two-component distribution model is shown for various mix ratios and relative humidities. At relative humidities equal to 0.8 and above, β decreases with the mix ratio. However, for relative humidities less than 0.8 this trend is reversed around 9 μm. This is due to the lower index of refraction of the continental aerosols.

Attempts have been made by Barnhardt and Streete [1] and Hodges [2] to compare the results obtained from their models with experimental values of the aerosol extinction coefficients measured by Yates and Taylor [11] and Kurnick, Zitter and Williams [12]. Unfortunately such comparisons cannot, at this moment, lead to definite conclusions because the experimental measurements of β so far have not been accompanied by information of the particle number distribution. By changing the parameters in an assumed distribution, one can obtain a range of different values for β. In fact, Curcio [13] has shown that different distributions can reproduce the same extinction coefficients. However, it is of interest to compare the present results with those in [1] and [2] based on a simplified model.

In general, the values of β obtained in [1] are higher. This is because the growth factor assumed by Barnhardt and Streete is larger than the empirical values we have adopted. Take, for example, the case where the mix ratio is 0.5, i.e. the continental and maritime components are in 1:1 ratio. The results of [1] are higher than those obtained here for all the relative humidities except the highest ($f = 0.99$). When $f = 0.99$, the agreement is reasonable in the 3-5 μm region. In the 8-15 μm region our results show a dip around 10 μm which is absent in [1]. In Fig. 2 we see that the growth factor used in [1] is between the empirical values for the continental and maritime components when $f = 0.99$. It can also be noted that the presence of a dip in the values of β in the 10-12 μm spectral region is characteristic for many fogs and clouds [14].

In [2], Hodges has assumed the same single growth factor as in [1] for both the continental and maritime components. He has also assumed that both components had the same refractive index except which now could be complex. The imaginary part of the refractive index has been taken to be equal to that of water. From Table II, we can readily see that the assumption $\eta''_o = \eta''_w$ leads to an overestimation of the aerosol absorption coefficient which becomes more serious in the longer wavelength region ($\lambda \gtrsim 10$ μm). This is particularly clear for the maritime component. In fact, the values of β obtained in [2] for the 3-5 μm region are substantially the same as those in the 8-15 μm region. From our results, β is smaller in the latter region.

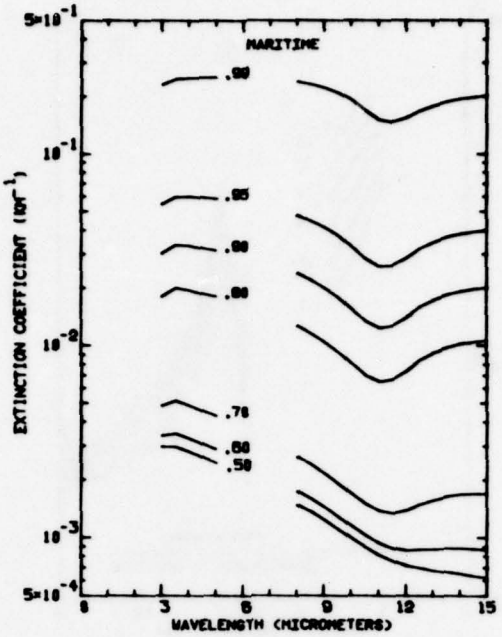

FIGURE 3 - Extinction coefficient as a function of wavelength for maritime aerosols. The number accompanying each curve is the relative humidity.

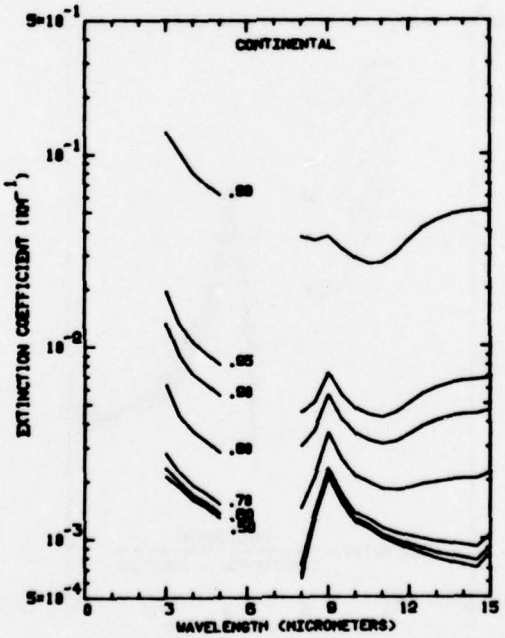

FIGURE 4 - Extinction coefficient as a function of wavelength for continental aerosols. The number accompanying each curve is the relative humidity.

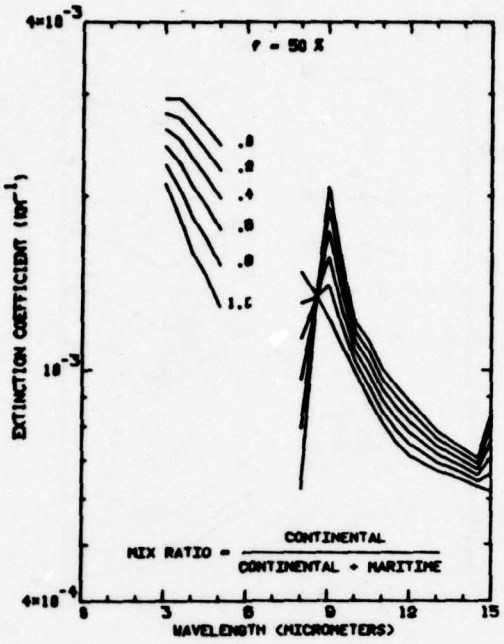

FIGURE 5 - Extinction coefficient as a function of wavelength for different mix ratios. The number accompanying each curve is the mix ratio.

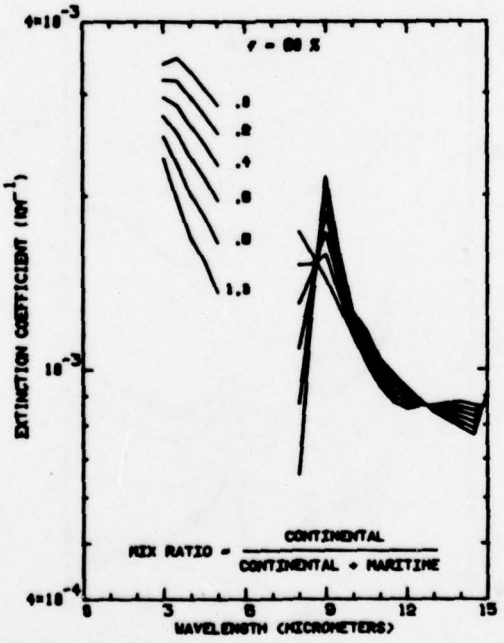

FIGURE 6 - Extinction coefficient as a function of wavelength for different mix ratios. The number accompanying each curve is the mix ratio.

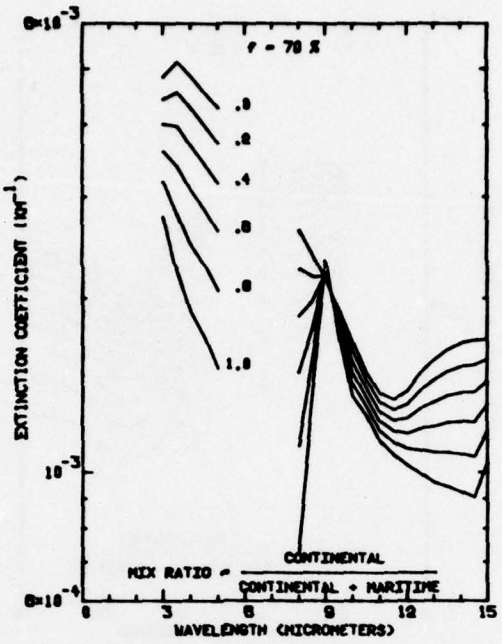

FIGURE 7 - Extinction coefficient as a function of wavelength for different mix ratios. The number accompanying each curve is the mix ratio.

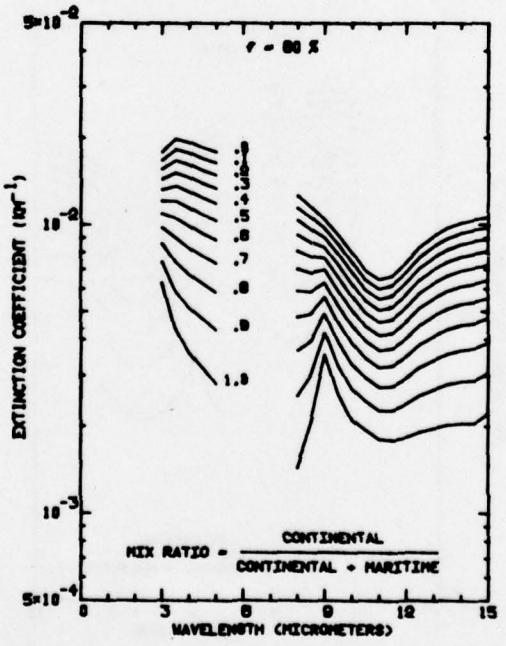

FIGURE 8 - Extinction coefficient as a function of wavelength for different mix ratios. The number accompanying each curve is the mix ratio.

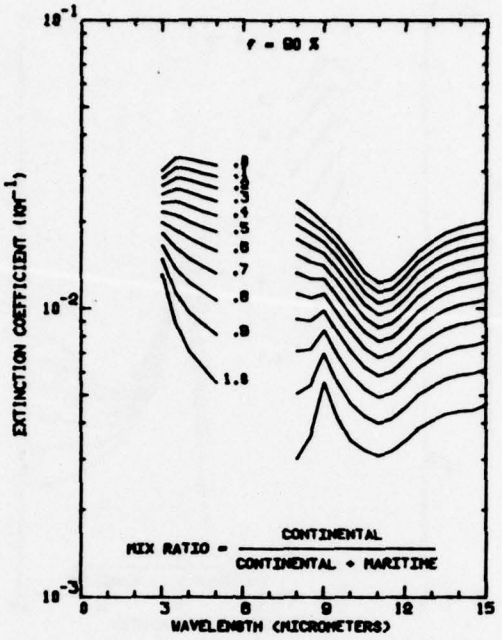

FIGURE 9 - Extinction coefficient as a function of wavelength for different mix ratios. The number accompanying each curve is the mix ratio.

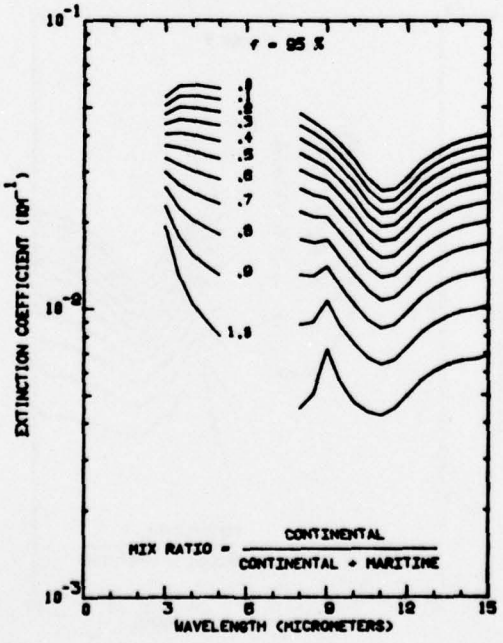

FIGURE 10 - Extinction coefficient as a function of wavelength for different mix ratios. The number accompanying each curve is the mix ratio.

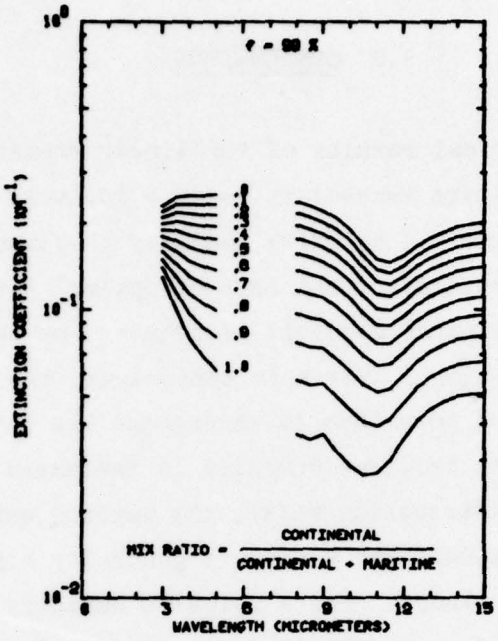

FIGURE 11 - Extinction coefficient as a function of wavelength for different mix ratios. The number accompanying each curve is the mix ratio.

5.0 CONCLUSIONS

Based on empirical results of the linear mass-increase coefficients and the dry-state aerosol refractive indices, we have studied the influence of increase in relative humidity on aerosol extinction. In doing so, we have used different sets of optical parameters for the maritime and continental aerosols as required by the difference in their chemical compositions. For both components, the values of β increase by a factor of more than 10 throughout the 3-5 and 8-15 μm window regions when the relative humidity is increased from 0.5 to 0.99. For a two-component distribution model, the aerosol extinction coefficient in the shorter wavelength window is generally higher than that in the longer wavelength window. For a relative humidity higher than 0.8, a dip in the value of β around 10-11 μm is also noticed.

In contradistinction to the results previously obtained by using simpler models, the influence of relative humidity on aerosol extinction has shown to be more dramatic and a better aerosol transmission condition in the 8-15 μm region is also indicated.

6.0 ACKNOWLEDGEMENTS

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CRDV R-4121/79 (NON CLASSIFIE)

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"The effects of relative humidity on atmospheric aerosol extinction in the 3-5 μm and 8-15 μm spectral regions"
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The variation of the infrared aerosol extinction coefficient as a function of relative humidity is studied. This is based on a two-component particle distribution model. The aerosol extinction coefficient increases by a factor of more than 10 throughout the 3-5 and 8-15 μm spectral regions as the relative humidity increases from 0.5 to 0.99. For high relative humidities, a region of lower extinction around 10-11 μm is noticed. (U)

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