

Approximation for the absorption coefficient of airborne atmospheric aerosol particles in terms of measurable bulk properties

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ABSTRACT

The absorption coefficient of airborne atmospheric aerosol particles can be approximated by

$$\frac{12\pi}{\lambda} \cdot \frac{k}{\rho} \cdot \frac{1}{n^2 + 2} \frac{M}{V_k}$$

where λ is the wavelength of radiation, $n - ik$ is the mean complex refractive index, ρ the mean bulk density, and M/V_k the mass of the particles per unit volume of air. This approximation gives good results at relative humidities between 0 and 0.95 for the wavelengths of radiation between $0.55 \mu\text{m}$ and $2.0 \mu\text{m}$ and between $9.25 \mu\text{m}$ and $12.0 \mu\text{m}$. Basing on this approximation it is possible to determine the single scattering albedo of airborne atmospheric aerosol particles with known measuring techniques.

1. Introduction

In the last few years the influence on radiative heating and cooling of the atmospheric air by aerosol particles has become an important topic (e.g. Yamamoto & Tanaka, 1972, Eschelbach, 1973, Grassl, 1973, 1974). Beyond that the influence of radiation on meteorological parameters near the ground has been investigated (e.g. Zdunkowski & McQuage, 1972). The treatment of these problems requires among other information the knowledge of the absorption coefficient of the airborne atmospheric aerosol particles as functions of the wavelength of electromagnetic radiation—especially in the visible and near infrared wavelength region, the relative humidity, and the aerosol type. In the present paper a possibility will be discussed, how the absorption coefficient of airborne atmospheric aerosol particles can be determined experimentally. This discussion bases on an approximation to the theory of scattering and absorption of electromagnetic radiation by homogeneous spheres (Mie, 1908).

2. Approximation formula for the absorption coefficient

The absorption coefficient σ_A of N_K homogeneous spheres within a volume of air V_K is given by

$$\sigma_A = \frac{1}{V_K} \sum_{j=1}^{j=N_K} \pi r_j^2 \kappa_{Aj}(\lambda, r_j, n_j, k_j) \quad (1)$$

λ is the wavelength of radiation, r_j is the radius, κ_{Aj} the efficiency factor of absorption and $n_j - ik_j$ the complex refractive index of the j th sphere. It is assumed independent and single scattering.

From Penndorf (1962) it can be derived an approximation formula for the efficiency factor of absorption κ_A being valid in the range $1.25 \leq n \leq 1.75$ and $k \leq 1$ of the complex refractive index:

$$\kappa_A \approx \frac{24 n k \alpha}{(n^2 + 2)^2 + k^2(2n^2 + k^2 - 4)} + \dots$$

with $\alpha = \frac{2\pi r}{\lambda} \leq 0.8$ (2)

α is called the size parameter. Within the range of validity of eq. (2) it can be approximated further

$$\frac{n}{(n^2 + 2)^2 + k^2(2n^2 + k^2 - 4)} \approx \frac{1}{3(n^2 + 2)}$$

leading to

$$\kappa_A \approx \frac{8ka}{n^2 + 2} + \dots \quad (3)$$

To facilitate the following considerations the generalized size parameter of absorption α_A is defined by

$$\alpha_A = \frac{8ka}{n^2 + 2} \quad (4)$$

This definition has been chosen as a tool for the classification of the values of the efficiency factor of absorption $\kappa_{A,MIE}$ that is computed from the ex-

act Mie-theory. A justification of this choice is given in Fig. 1, where $\kappa_{A,MIE}$ is plotted as a function of α_A . There have been chosen only those complex refractive indices from the range of validity of the eqs. (2) and (3). It can be seen easily that the approximation

$$\kappa_{A,MIE} \approx \alpha_A \quad \text{for} \quad \alpha_A \leq 1$$

and

$$\kappa_{A,MIE} \approx 1 \quad \text{for} \quad \alpha_A > 1 \quad (5)$$

describes the curves very well for all complex refractive indices used. The best results would be obtained from $\kappa_{A,MIE} \approx \alpha_A$ in the range $\alpha_A \leq k$, i.e. $\alpha \leq (n^2 + 2)/8$. However this is an admissible restriction for α . It will be shown in Table 2, that the approximation $\kappa_{A,MIE} \approx \alpha_A$ can be used in the range $\alpha_A \leq 1$, i.e. $\alpha \leq (n^2 + 2)/(8k)$. Thus the upper

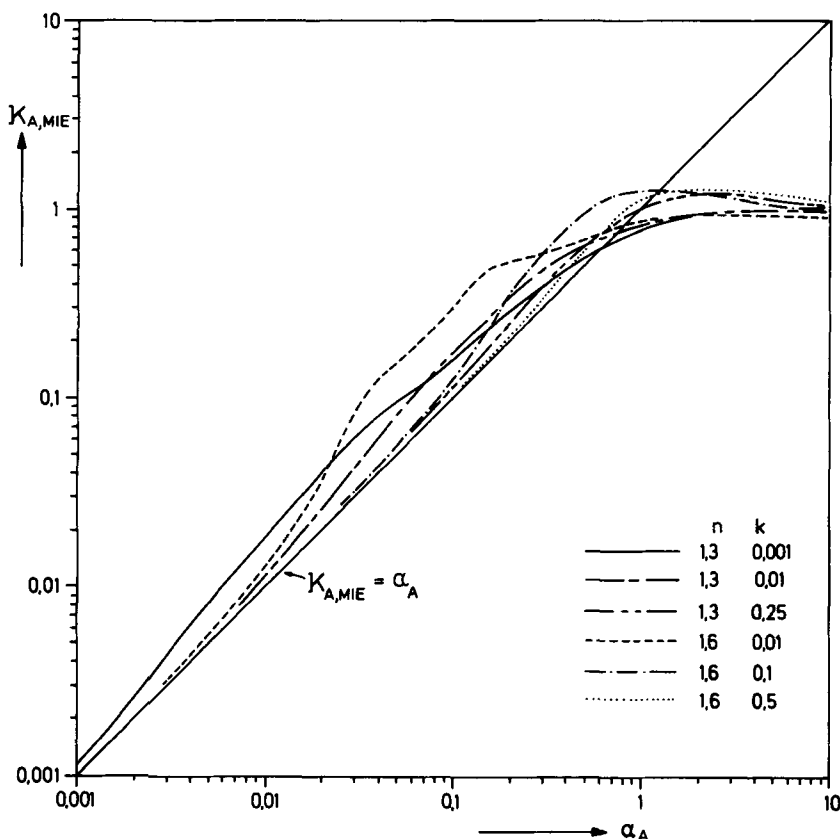


Fig. 1. Efficiency factor of absorption for a homogeneous sphere $\kappa_{A,MIE}$ after Mie (1908) versus the generalized size parameter of absorption α_A for several complex refractive indices $n - ik$ of the sphere.

limit of tolerable α -values increases when k decreases.

The approximation (5) is applicable to atmospheric aerosol particles since there have been used the usual mean complex refractive indices the atmospheric aerosol particles attain in the whole range of relative humidity and at wavelengths of radiation between $0.55 \mu\text{m}$ and $12.0 \mu\text{m}$ (Eiden, 1966; Hänel, 1968, 1972a; Fischer, 1971, 1973, 1975; Volz, 1973).

Using the approximation (5) the absorption coefficient σ_A is

$$\begin{aligned} \sigma_A &\cong \frac{1}{V_K} \left[\sum_{j=1}^{\tilde{N}} \pi r_j^2 \alpha_{Aj} + \sum_{j=\tilde{N}+1}^{N_K} \pi r_j^2 \right] \\ &= \frac{1}{V_K} \left[\frac{12\pi}{\lambda} \sum_{j=1}^{\tilde{N}} \frac{k_j}{n_j^2 + 2} (\frac{4}{3}\pi r_j^3) \right. \\ &\quad \left. + \sum_{j=\tilde{N}+1}^{N_K} \pi r_j^2 \right] \end{aligned} \quad (6)$$

where $\tilde{N}$ is the number of particles with a generalized size parameter of absorption $\alpha_A \leq 1$. Thus we have two regions of absorption types:

- In the first region, when $\alpha_A \leq 1$, the absorption of each sphere is proportional to the product of its volume $\frac{4}{3}\pi r_j^3$ with the imaginary part k_j of its refractive index.
- In the second region, when $\alpha_A > 1$, the absorption of each sphere is equal to its geometrical cross section and therefore independent from the refractive index.

The assumption, that all the first $\tilde{N}$ spheres have the same complex refractive index $n - ik$ and the same bulk density ρ , leads to

$$\begin{aligned} \sigma_A &\cong \frac{1}{V_K} \left[\frac{12\pi}{\lambda} \frac{k}{\rho} \frac{1}{n^2 + 2} \sum_{j=1}^{\tilde{N}} m_j \right. \\ &\quad \left. + \sum_{j=\tilde{N}+1}^{N_K} \pi r_j^2 \right] = \frac{12\pi}{\lambda} \frac{k}{\rho} \frac{1}{n^2 + 2} \frac{\tilde{M}}{V_K} \\ &\quad + \frac{\tilde{Q}}{V_K} \end{aligned} \quad (7)$$

where $m_j = \frac{4}{3}\pi r_j^3 \rho$ is the mass of the j th sphere, $\tilde{M} = \sum_{j=1}^{\tilde{N}} m_j$ and $\tilde{Q} = \sum_{j=\tilde{N}+1}^{N_K} \pi r_j^2$.

When the values of the generalized size parameter of absorption are small compared to 1 for all or the most important absorbing particles, it is

$$\begin{aligned} \sigma_A &\cong \frac{12\pi}{\lambda} \frac{k}{\rho} \frac{1}{n^2 + 2} \frac{M}{V} \\ \text{with } M &= \sum_{j=1}^{N_K} m_j \end{aligned} \quad (8)$$

M/V_K is the mass of all spheres per unit volume of air. The neglect of the second term of eq. (7) $\tilde{Q}/V_K$ is justified by the results from Table 2.

This latter equation is in agreement with the computational results for single particles by Plass (1966) and Bergstrom (1973); the first author stated a proportionality between σ_A and k for the visible wavelength region. Recently Waggoner et al. (1973) have derived a formula reading

$$\sigma_A \cong F(\lambda, n) \frac{k}{\rho} \frac{M}{V_K}$$

but they did not specify $F(\lambda, n)$. Comparison with eq. (8) gives $F(\lambda, n) = 12\pi/(n^2 + 2) \cdot \lambda$. The approximation (8) is the basis for the subsequent considerations, since the second part of eq. (7) cannot be determined from size distribution measurements with an accuracy justifying the formal complication of this equation. The latter equation, indicating a proportionality between the absorption coefficient of the airborne particles and the mass (volume) of the particles per unit volume of air, is of special interest. It is an outstanding feature of eq. (8), that within the range of its validity the absorption coefficient is independent from the shape of the aerosol size frequency distribution.

3. Comparison of equation (8) with Mie's theory by model computations

The validity of the approximation (8) for the absorption coefficient has been proved by computations. To approach reality they are based upon measured aerosol properties. The latter ones have been obtained from measurements on samples of a large number of aerosol particles being collected with a jet impactor. The experimental data have

been described in detail by Hänel (1972a, b), where all the appertaining references are listed. Additionally there have been deduced mean complex refractive indices of dry aerosol particles for $\lambda > 1.0 \mu\text{m}$ from the measurements of Volz (1972, 1973) as well as from Fischer (1973, 1975). For several wavelengths of radiation between $0.55 \mu\text{m}$ and $12.0 \mu\text{m}$ and for relative humidities up to 0.99 computations are performed for the following three aerosol types (Hänel 1972a, b):

Model 3: Maritime aerosol over the Atlantic, April 1968

Model 5: Urban aerosol in January 1970 at Mainz, Germany

Model 6: Clean air aerosol at Hohenpeissenberg 1000 m above mean sea level in summer 1970, Germany.

The relevant aerosol size frequency distributions of the dry particles are given in Fig. 2. The appertaining complex refractive indices of water and mean complex refractive indices of the dry spheres are compiled in Table 1.

On the one hand values $\sigma_{A,MIE}$ of the absorption coefficient have been computed by the Mie-theory. These are compared with those values σ_A of the absorption coefficient from formula (8). Generally it has been assumed that in a dry state all particles have the same chemical composition and structure independent from their size. Therefore the mean bulk properties obtained from samples can be used. Moreover for the Mie-computations the effect of curvature on the equilibrium vapour pressure of water over the surface of aerosol parti-

cles has been incorporated. Thus at the highest relative humidities smaller particles show a smaller radius increase due to water vapour condensation than larger ones. For this reason the course with

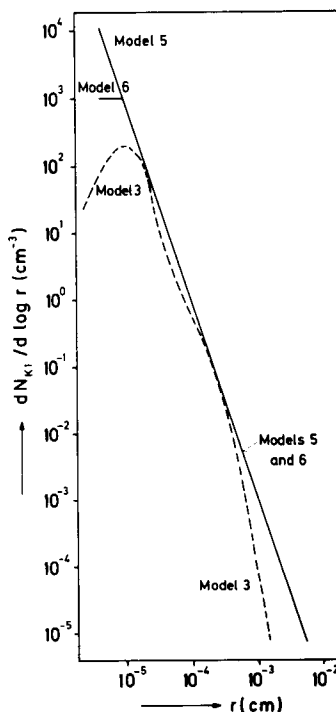


Fig. 2. Size frequency distributions of dry aerosol particles for the aerosol models 3, 5 and 6 used during computations. $N_{K1} = N_K/V_K$ is the particle number per unit volume of air, r is the equivalent radius, i.e. the radius of a sphere with the particles volume.

Table 1. Complex refractive index $n_w - ik_w$ of water and mean complex refractive index $n_0 - ik_0$ of the dry aerosol substance for the models 3, 5 and 6 at the wavelengths of radiation considered

		$\lambda [\mu\text{m}]$					
		0.55	2.0	3.0	9.25	10.0	12.0
Model 3	n_w	1.334	1.304	1.315	1.257	1.214	1.11
	k_w	0	0.00108	0.259	0.0422	0.0532	0.244
	n_0	1.55	1.47	1.338	1.65	1.75	1.75
	k_0	0.055	0.132	0.264	0.528	0.198	0.198
Model 5	n_0	1.55	1.466	1.33	1.55	1.7	1.7
	k_0	0.048	0.136	0.272	0.544	0.34	0.204
Model 6	n_0	1.51	1.472	1.432	1.6	1.7	1.7
	k_0	0.015	0.04	0.08	0.16	0.06	0.06

relative humidity of the mean complex refractive index is dependent slightly from the particle size in dry state. All these dependencies have been neglected evaluating the approximation (8). There have been introduced the dependencies from relative humidity of the real and of the imaginary part of the mean complex refractive index, of the mean bulk density, and of the mass which have been measured on the relevant aerosol samples. It has been shown in the past (Hänel, 1970, 1971) that this procedure gives good values as long as the relative humidity remains smaller than about 0.9 to 0.95.

The results of the model computations are compiled in Table 2, where the percentage deviation

$100 \cdot (\sigma_A / \sigma_{A,MIE} - 1)$ of the absorption coefficient σ_A computed with eq. (8) from the absorption coefficient $\sigma_{A,MIE}$ computed with Mie-theory are given. Those wavelengths of radiation that are important for the estimation of aerosol effects on the radiative transfer within the atmosphere have been used. The wavelength $9.25 \mu\text{m}$ used is of special interest, since there often can be formed a strong aerosol absorption. The computational values are valid only for increasing relative humidity from the dry state $f=0$ on. Hysteresis effects at medium relative humidities, producing a larger water uptake at decreasing—compared to that at increasing relative humidity, are not considered here, since they do not alter the results in principle. The values

Table 2. Percent deviation $100 \cdot (\sigma_A / \sigma_{A,MIE} - 1)$ of the absorption coefficient σ_A computed with equation (8) from the absorption coefficient $\sigma_{A,MIE}$ computed with Mie-theory versus relative humidity f and wavelength λ of radiation

		$\lambda \mu\text{m}$					
		0.55	2.0	3.0	9.25	10.0	12.0
		$100 \cdot (\sigma_A / \sigma_{A,MIE} - 1)$					
Model 3	0	-10	1	29	1	-28	-29
	0.2	-9	1	29	1	-28	-28
	0.4	-10	0	30	1	-28	-28
	0.6	-11	-1	32	0	-27	-24
	0.7	-13	-7	42	-1	-20	-9
	0.8	-23	-20	51	-6	-16	6
	0.9	-28	-26	62	-8	-13	15
	0.95	-32	-31	76	-9	-10	25
	0.99	-40	-38	135	-7	0	56
Model 5	0	-5	15	40	20	1	-13
	0.2	-5	14	40	20	1	-13
	0.4	-6	14	40	19	1	-12
	0.6	-7	12	52	20	4	4
	0.7	-9	7	45	16	1	1
	0.8	-15	-2	53	12	2	14
	0.9	-18	-7	81	14	11	42
	0.95	-25	-17	83	8	8	42
	0.99	-37	-27	144	13	21	80
Model 6	0	-29	-20	-5	-13	-38	-37
	0.2	-29	-20	-4	-13	-37	-36
	0.4	-29	-20	-2	-13	-37	-35
	0.6	-29	-21	2	-12	-35	-30
	0.7	-29	-20	18	-10	-30	-16
	0.8	-31	-24	29	-12	-24	-5
	0.9	-33	-25	65	-3	-7	27
	0.95	-34	-28	64	-8	-10	24
	0.99	-41	-33	135	4	11	70

from Table 2 show, that for all wavelengths of radiation except $3.0\ \mu\text{m}$, the approximation (8) gives errors smaller than 35 to 40% at relative humidities smaller than 0.95.

These somewhat surprising results shall now be explained:

1. $\lambda = 0.55\ \mu\text{m}$ to $2.0\ \mu\text{m}$:

(a) Dry state: The generalized size parameters α_A of absorption for the largest particles range between approximately 6 and 20. Therefore we have one region of α_A , where the approximation for the efficiency factor of absorption $\kappa_A \cong \alpha_A$ gives too small values, i.e., at $\alpha_A \leq 1$, and another region, where this approximation gives too large values, i.e., at $\alpha_A > 1$. Finally the errors of a different sign involved in the approximation $\kappa_A \cong \alpha_A$ balance out each other to a high degree.

(b) Large relative humidities: The generalized size parameters of absorption of practically all particles are smaller than 1, due to the large decrease of the imaginary part of refractive index. Therefore the counterbalance described above is more deficient giving rise to more negative errors.

2. $\lambda = 3.0\ \mu\text{m}$:

(a) Dry state: The α_A -values of the largest particles are considerably larger than one. For models 3 and 5 the imaginary parts of the refractive indices are very large. Thus the approximation $\kappa_A \cong \alpha_A$ is very good for $\alpha_A \leq 1$ and gives too large values for $\alpha_A \geq 1$. This leads to large positive errors for α_A . For the model 6 the case 1a is valid.

(b) Large relative humidities: Since the imaginary part of refractive index remains nearly constant (models 3 and 5) or increases strongly (model 6) with relative humidity, the α_A -values have become very large at large relative humidities. Thus large positive errors are found.

3. $\lambda = 9.25\ \mu\text{m}$: In the whole range of relative humidity case 1a is applicable to all models.

4. $\lambda = 10\ \mu\text{m}$: To the models 3 and 5 in dry state, the case 1a is applicable. The approximation $\kappa_A \cong \alpha_A$ is valid for practically all particles for model 6 in the whole range of relative humidities considered here and for models 3 and 5 at large relative humidities.

5. $\lambda = 12\ \mu\text{m}$: In dry state case 1a is applicable to

all models, and at large relative humidities we find for all models the same situation which has been described in case 2a for the models 3 and 5.

These examples show, that in the visible and near infrared wavelength region as well as at wavelengths around $10\ \mu\text{m}$ the approximation (8) gives good results. The discussion yielded, that in most cases the eq. (8) is not a good approximation because $\kappa_A \cong \alpha_A$ is valid for all particles. Much more the validity of eq. (8) is based upon a counterbalance of errors of different sign involved in $\kappa_A \cong \alpha_A$ for the largest particles within the aerosol size frequency distribution.

Thus the approximation (8) can be applied only to samples of atmospheric suspensions because of their broad size frequency distribution. Thus, for the determination of the absorption coefficient of collectives of airborne spheres no information on their size frequency distribution is necessary. Only their mass per unit volume of air as well as their complex refractive index and their bulk density must be known as the determining properties besides the wavelength of radiation.

4. Discussion

In the previous section we have presupposed indirectly the validity of the Mie-theory for atmospheric aerosol particles. Since at all relative humidities they are not homogeneous spheres, MIE-theory is not obviously applicable. However, Medalia & Richards (1972) have measured the efficiency factors of absorption of agglomerates of carbon spheres in the visible wavelength region and could verify Mie-theory. Moreover our present picture of the formation and of the size frequency distribution of atmospheric aerosol particles (e.g. Whitby, 1974) tells that the most efficiently absorbing particles usually should be agglomerates with volume equivalent radii between $0.05\ \mu\text{m}$ and $1\ \mu\text{m}$. Therefore the model computations presented can be regarded as reliable. But it must not be forgotten that there are some open questions, i.e. when the most efficiently absorbing particles are large, not agglomerated and mineral.

Secondly the approximation (8) includes an improvement for computing the mean complex refractive index of mixed particles or samples of

particles. This arises from taking into account the influence of the real part of the refractive index on the absorption of radiation. It should be used when the most reliable averaging for computation of the absorption of electromagnetic radiation is desired. Comparison of the first sum of eq. (6) with (8) gives

$$\frac{k}{n^2 + 2} = \sum_i \frac{k_i}{n_i^2 + 2} \frac{V_i}{V} \quad (9)$$

where $n - ik$ and V are the mean complex refractive index and the bulk volume of a particle or a sample and $n_i - ik_i$ and V_i are the refractive indices and the bulk volumes of the pure components before mixing and/or agglomeration. Together with the formula for the real part of the mean refractive index $n = \sum_i n_i V_i / V$ (Hänel, 1968) $n - ik$ can be computed. Until now—the model computations of this paper included—the formula $k = \sum_i k_i V_i / V$ has been used. This formula meets eq. (9), when the n_i and n are almost equal. Thus eq. (9) gives an improvement, when n_i and n are not equal. The range of validity of eq. (8) is $1.25 \leq (n, n_i) \leq 1.75$ and $0 \leq (k, k_i) \leq 1$.

5. Conclusions

The computational results indicate, that the absorption coefficient of samples of airborne spheres or aerosol particles containing the whole size spectrum is proportional to the particles mass per unit volume of air, the imaginary part of their mean refractive index divided by their mean bulk density, and to a function of the real part of their mean refractive index (eq. 8). All these properties can be determined by measurement (Fischer, 1971, 1973, 1975; Hänel, 1968). It is a facilitation, that no information on the size frequency distribution of

the particles is necessary. The approximation (8) allows the computation of the single scattering albedo ω of a sample of airborne aerosol particles with measurable properties, when not the highest accuracy is expected:

$$\omega = 1 - \frac{\sigma_A}{\sigma_E} \cong 1 - \frac{12\pi}{\lambda} \frac{k}{\rho} \frac{1}{n^2 + 2} \frac{M}{V_K \sigma_E} \quad (12)$$

σ_E is the measurable extinction coefficient of the airborne particles.

The discussion yields, that the approximation (8) for the absorption coefficient leads to the more elaborate set of equations for the mean refractive index of a mixed particle or a sample. This new formula is applicable when the absorption coefficient shall be computed. It is an open question what set of refractive index mixture rules is the most reliable one, e.g. for the computation of the extinction coefficient or the scattering function. Moreover it is not sure, whether different mixture rules are necessary for very small and for very large particles compared to the wavelength of radiation considered. In the case of the absorption coefficient, an arbitrary mixture rule could be used for the very large particles since their efficiency factors of absorption are practically independent from the complex refractive index regarding atmospheric conditions (i.e. $1 \leq n \leq 2$, $0 \leq k \leq 1$). All these problems are far from being completely understood and thus have to be investigated in the future.

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АППРОКСИМАЦИЯ КОЭФИЦИЕНТА ПОГЛОЩЕНИЯ ЧАСТИЦ АТМОСФЕРНОГО АЭРОЗОЛЯ С ПОМОЩЬЮ ИЗМЕРЯЕМЫХ ОБЪЕМНЫХ ПАРАМЕТРОВ

Коэффициент поглощения взвешенных частиц атмосферного аэрозоля может быть аппроксимирован выражением $\frac{12\pi k}{\lambda} \frac{1}{\rho n^2 + 2} \frac{M}{V_k}$, где λ — длина волны радиации, $n - ik$ — комплексный показатель преломления, ρ — средняя объемная плотность и M/V_k — масса частиц на единицу

объема воздуха. Это приближение дает хорошие результаты при относительных влажностях между 0 и 0,95 для длин волн радиации между 0,55 и 2,0 мкм и между 9,25 и 12,0 мкм. На основе этого приближения с известной техникой измерений можно определить альбедо единичного рассеяния взвешенных частиц атмосферного аэрозоля.