The Single-Scattering Albedo of Atmospheric Aerosol Particles as a Function of Relative Humidity

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1. Introduction

The general circulation of the atmosphere is affected by the radiative heating and cooling due to the aerosol particles. In this context the single-scattering albedo ω of atmospheric aerosol particles is a property of immediate interest (compare Chandrasekhar, 1950; Yamamoto and Tanaka, 1972; Eschelbach, 1973; Grassl, 1973, 1974). It is defined by

$$\omega = 1 - \sigma_A/\sigma_E, \tag{1}$$

where σ_A is the absorption coefficient and σ_E the extinction coefficient of the atmospheric aerosol particles.

For electromagnetic radiation penetrating a volume of air containing particles, ω is the fraction of attenuated electromagnetic radiation scattered by the particles. The remaining fraction $1-\omega$ is transformed into another form of energy or of electromagnetic radiation of another wavelength. Since ω is defined by the ratio of the absorption and the extinction coefficient, it is independent of the number of particles per unit volume. Only the shape of the size-frequency distribution (the ratios of the particle sizes to the wavelength of radiation), the mean complex refractive indices (the shapes), and the structures of the particles have influences. Condensation of water vapor takes place on the aerosol particles when the relative humidity increases, whereas water evaporates from the particles with decreasing relative humidity. Thus the sizes, the mean complex refractive indices of the particles, and consequently the single-scattering albedo are functions of relative humidity.

In this note computational results on the single-scattering albedo for visible and infrared radiation are presented as a function of relative humidity. The calculations are based on Mie theory (1908) for the scattering and absorption of electromagnetic radiation by homogeneous spheres. In making such computations, it is recognized that atmospheric aerosol particles are not homogeneous spheres at any relative humidity. It is presupposed only that Mie theory allows reliable computation of the single-scattering albedo. This has been justified by Blättner and Wells (1973) who have compared computed efficiency factors of extinction and absorption of homogeneous spheres and of water-coated homogeneous spheres and by the measurements of Medalia and Richards (1972) on the efficiency factors of absorption of agglomerates of carbon spheres.

2. Description of the models

The earlier model computations (Hänel, 1972a,b) used six types of atmospheric aerosol particles. The computations of this paper cover only the three most important ones among them at selected wavelengths of radiation between 0.55 and 12.0 μm:

Model 3: Maritime aerosol over the Atlantic, 13–16 April 1969.
Model 5: Urban aerosol at Mainz, January 1970.
Model 6: Clean air aerosol on top of the Hohenpeissenberg, elevation 1000 m MSL, in summer 1970.

(These model numbers agree with those in the earlier papers.) The size distributions pertaining to each of these models are given in Fig. 1.

The model calculations have the following experimental bases:

1) Measured aerosol size frequency distributions (Junge, 1963; Jaenicke *et al.*, 1971).

2) Measurements of the water uptake of samples of aerosol particles at different relative humidities (Hänel, 1972c). [Whenever the relative humidity was increasing or decreasing prior to a measurement, sufficient time was taken for thermodynamic equilibrium to set in.]

3) Related data of the mean densities and the real parts of the mean complex refractive index of samples of dry aerosol particles derived from measurements (Hänel, 1972c).

4) Imaginary parts of the mean complex refractive index of samples of dry aerosol particles (Volz, 1972, 1973; Fischer, 1973, 1975).

The model calculations are based upon the assumptions that (i) the particles are in thermodynamic equilibrium with the ambient moist air; (ii) particles are neither added to nor removed from the sample, and no coagulation occurs, when the relative humidity changes; (iii) the particles are assumed to be homogeneous spheres; and (iv) the dry particles are chemically and structurally uniform. Then the data on

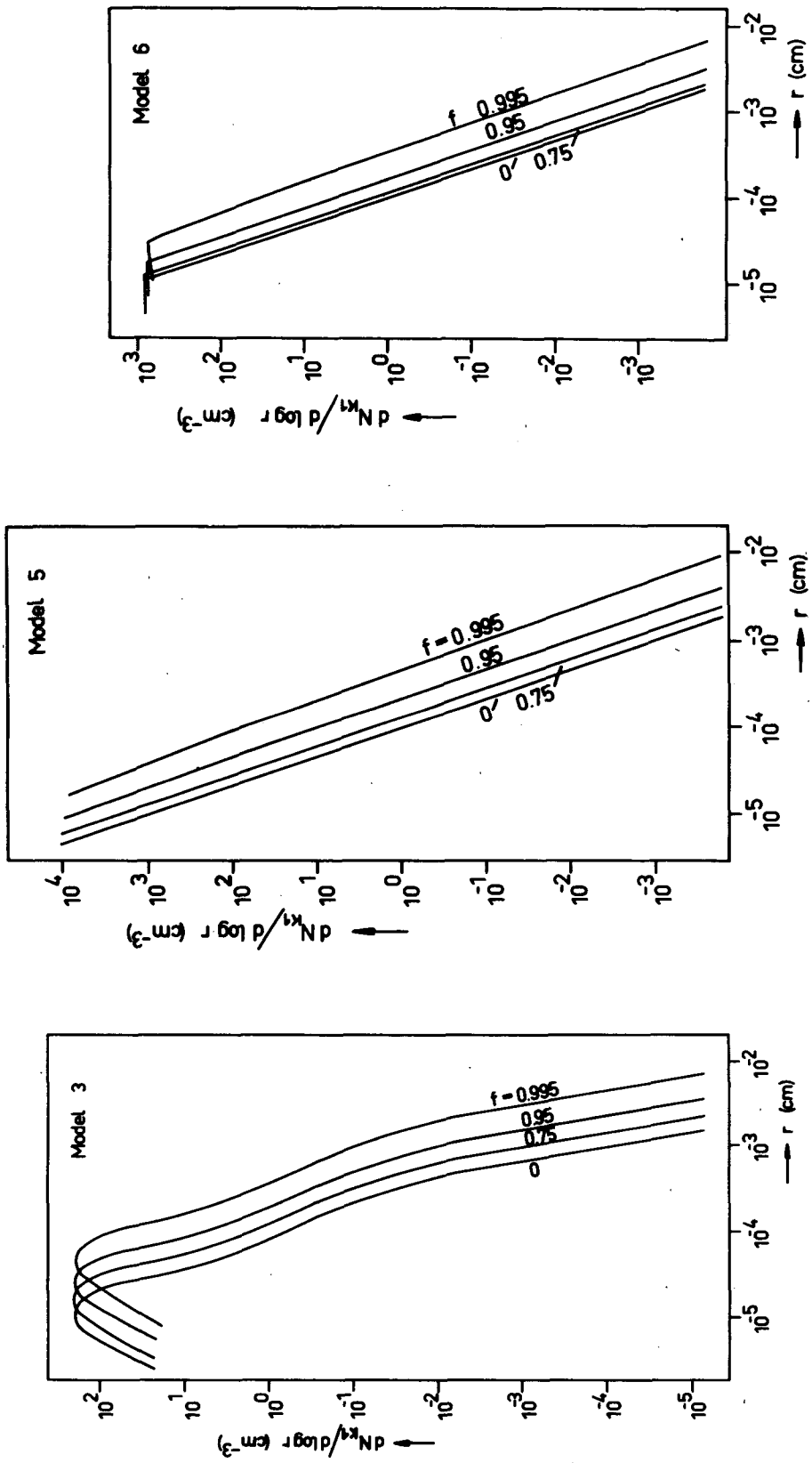

FIG. 1. Aerosol size distribution $dN_{K1}/d\log r$ vs relative humidity f, where N_{K1} is the particle number cm^{-3} and r the equivalent radius of the particles. The curves for $f = 0.75$ are valid for increasing relative humidity.

TABLE 1. Single-scattering albedo ω of atmospheric aerosol particles as a function of relative humidity f and wavelength λ of radiation together with the complex refractive indices $n_w - ik_w$ of water and mean complex refractive indices $n_0 - ik_0$ of the dry aerosol particles.

	Model 3 Wavelength λ (μm)						Model 5 Wavelength λ (μm)						Model 6 Wavelength λ (μm)					
f	0.55	2.0	3.0	9.25	10.0	12.0	0.55	2.0	3.0	9.25	10.0	12.0	0.55	2.0	3.0	9.25	10.0	12.0
	$\omega(f)$						$\omega(f)$						$\omega(f)$					
							Increasing relative humidity											
0	0.739	0.467	0.279	0.224	0.438	0.414	0.732	0.427	0.224	0.163	0.254	0.347	0.877	0.715	0.543	0.400	0.666	0.666
0.2	0.739	0.467	0.279	0.224	0.438	0.414	0.723	0.433	0.224	0.163	0.254	0.341	0.878	0.718	0.539	0.406	0.663	0.656
0.4	0.747	0.472	0.279	0.224	0.432	0.397	0.728	0.433	0.224	0.163	0.247	0.328	0.879	0.721	0.525	0.406	0.663	0.640
0.6	0.753	0.487	0.285	0.231	0.438	0.377	0.753	0.462	0.236	0.163	0.261	0.289	0.884	0.728	0.495	0.406	0.657	0.584
0.65	0.773	0.516	0.285	0.231	0.433	0.334	0.770	0.488	0.235	0.170	0.260	0.259	0.885	0.731	0.482	0.406	0.654	0.566
0.7	0.801	0.562	0.296	0.249	0.422	0.273	0.790	0.513	0.238	0.184	0.260	0.234	0.886	0.733	0.479	0.411	0.651	0.561
0.75	0.858	0.674	0.306	0.289	0.441	0.191	0.824	0.577	0.248	0.199	0.279	0.195	0.902	0.763	0.416	0.420	0.626	0.426
0.8	0.880	0.725	0.313	0.320	0.443	0.174	0.850	0.626	0.254	0.223	0.293	0.170	0.921	0.805	0.371	0.463	0.592	0.303
0.85	0.891	0.750	0.323	0.341	0.447	0.167	0.871	0.668	0.255	0.246	0.300	0.156	0.932	0.830	0.360	0.451	0.568	0.250
0.9	0.910	0.794	0.339	0.382	0.456	0.158	0.890	0.705	0.262	0.264	0.311	0.139	0.939	0.842	0.348	0.457	0.554	0.227
0.95	0.935	0.854	0.351	0.451	0.469	0.159	0.932	0.816	0.288	0.352	0.361	0.137	0.954	0.881	0.342	0.485	0.528	0.187
0.975	0.954	0.904	0.363	0.516	0.489	0.159	0.959	0.887	0.311	0.436	0.399	0.136	0.969	0.924	0.349	0.519	0.502	0.169
0.99	0.967	0.944	0.398	0.572	0.507	0.177	0.979	0.939	0.330	0.514	0.442	0.148	0.984	0.952	0.365	0.558	0.489	0.168
0.995	0.979	0.959	0.420	0.604	0.526	0.193	0.987	0.956	0.350	0.555	0.470	0.163	0.989	0.964	0.384	0.580	0.496	0.178
							Decreasing relative humidity											
0.2	0.742	0.472	0.279	0.224	0.432	0.403	0.728	0.433	0.224	0.163	0.254	0.334	0.879	0.721	0.517	0.400	0.663	0.627
0.4	0.746	0.477	0.286	0.224	0.443	0.392	0.741	0.448	0.224	0.163	0.254	0.312	0.882	0.723	0.506	0.400	0.660	0.604
0.6	0.771	0.512	0.285	0.224	0.433	0.334	0.773	0.488	0.235	0.177	0.260	0.256	0.886	0.733	0.482	0.406	0.654	0.563
0.65	0.883	0.618	0.301	0.262	0.438	0.223	—	—	—	—	—	—	0.888	0.733	0.477	0.411	0.651	0.548
0.7	0.862	0.682	0.303	0.297	0.441	0.190	0.812	0.555	0.247	0.195	0.275	0.207	0.896	0.752	0.437	0.415	0.635	0.769
0.75	0.871	0.700	0.311	0.306	0.441	0.182	0.844	0.615	0.253	0.218	0.289	0.174	—	0.796	0.379	0.437	0.598	0.323
n_w	1.334	1.304	1.351	1.257	1.214	1.11	1.334	1.304	1.351	1.257	1.214	1.11	1.334	1.304	1.351	1.257	1.214	1.11
k_w	0	0.00108	0.259	0.0422	0.0532	0.244	0	0.00108	0.259	0.0422	0.0532	0.244	0	0.00108	0.259	0.0422	0.0532	0.244
n_0	1.55	1.47	1.34	1.65	1.76	1.75	1.55	1.47	1.33	1.55	1.7	1.7	1.51	1.47	1.43	1.60	1.7	1.7
k_0	0.055	0.132	0.264	0.528	0.198	0.189	0.048	0.136	0.272	0.544	0.34	0.204	0.015	0.04	0.08	0.16	0.06	0.06

samples of aerosol particles are applicable to airborne particles. The last assumption is justified because of the mixed structure of the particles in the atmosphere due to coagulation.

The effects of curvature on the equilibrium vapor pressures of water over the surfaces of aerosol particles have been incorporated, leading to a significantly smaller radius increase of the smallest particles compared to that of the largest particles especially at high relative humidities. Consequently the computer program allowed each airborne particle to have its own refractive index as a function of relative humidity.

Due to hysteresis effects the measured water uptakes by the samples are smaller for increasing than for decreasing relative humidity (see also Wall, 1942; Junge, 1952; Orr *et al.*, 1958; Winkler and Junge, 1972). Therefore the computations are performed for both increasing and decreasing relative humidity.

3. Results and conclusions

The results of the single-scattering albedo computations as a function of relative humidity f at selected wavelengths λ of radiation are compiled in Table 1 and summarized in Fig. 2. They allow the following conclusions.

1) At relative humidities below 0.6 the single-scattering albedo is nearly constant due to the small amounts of water condensed on the particles.

2) There are no large differences between the values for increasing and decreasing relative humidity. The differences are largest for the maritime aerosol model 3 at relative humidities around 0.65.

3) There is an increase of the single-scattering albedo of the particles with increasing relative humidity in most cases. In the region of short wavelengths, between approximately 0.3 and 2.5 μm, this increase of the single-scattering albedo is mostly due to a strong decrease of the imaginary part of the mean complex refractive index. Here the imaginary part of the complex refractive index of water is zero or very small compared to the imaginary part of the mean complex refractive index of the dry particles.

4) At a wavelength of 12.0 μm, and for model 6 at wavelengths of 3.0 and 10.0 μm, the single-scattering albedo first decreases with increasing relative humidity. Only for the highest relative humidities is an increase of the single-scattering albedo observed. This latter increase is due to the tendency of the single-scattering albedo to approach a value close to 0.5 (compare Deirmendjian 1969) The value 0.5 is approached when most of the particles have absorption efficiency factors close to 1 and extinction efficiency factors close to 2. These presuppositions are fulfilled at best at very high relative humidities where the sizes of the particles attain the largest values.

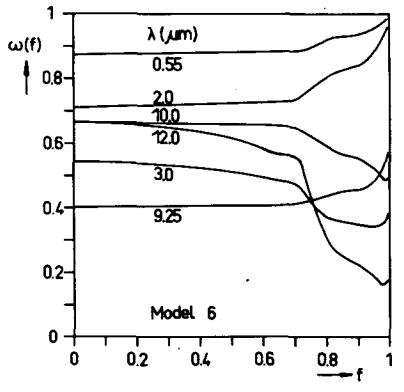

FIG. 2. Variation of the single-scattering albedo w with relative humidity f and wavelength λ for model 6.

The results show that the effect of relative humidity on the single-scattering albedo of atmospheric aerosol particles cannot be neglected when the relative humidity is larger than about 0.6. Thus a discussion of radiative transfer within the atmosphere must take this effect into account at those wavelengths of electromagnetic radiation for which the influence of the particles is significant.

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REFERENCES

Blättner, W. G., and M. B. Wells, 1973: Scattering characteristics of homogeneous and inhomogeneous particles: Scattering in the visible and near infrared. Sci. Rept AFCRL-TR-74-0025, Air Force Cambridge Research Laboratories, Bedford, Mass.

Chandrasekhar, S., 1950: *Radiative Transfer.* Clarendon Press, 1–12.

Deirmendjian, D., 1969: *Electromagnetic Scattering on Spheroid Polydispersions.* Elsevier, 290 pp.

Eschelbach, G., 1973: Computations of the influence of aerosols on the atmospheric radiation balance in the visible spectrum. *Beitr. Phys. Atmos.*, **46**, 249–261.

Fischer, K., 1973: Mass absorption coefficient of natural aerosol particles in the 0.4–2.4 μm wavelength interval. *Beitr. Phys. Atmos.*, **46**, 89–100.

——, 1975: Mass absorption indices of natural aerosol particles in the infrared. *Appl. Opt.*, **14**, 2851–2856.

Grassl, H., 1973: Aerosol influence on radiative cooling. *Tellus*, **25**, 386–395.

——, 1974: Atmospheric absorbers in the window-regions and their influence on radiative heating and cooling. *Proc. IAMAP/IAPSO Assembly*, Melbourne, Australia, January.

Hänel, G., 1972a: Computation of the extinction of visible radiation by atmospheric aerosol particles as a function of the relative humidity, based upon measured properties. *Aerosol Sci.*, **3**, 377–386.

——, 1972b: The ratio of the extinction coefficient to the mass of atmospheric aerosol particles as a function of the relative humidity. *Aerosol Sci.*, **3**, 455–460.

——, 1972c: Messung und Berechnung physikalischer Eigenschaften atmosphärischer Schwebeteilchen als Funktion der relativen Luftfeuchtigkeit. Dissertation, Johannes Gutenberg-Universität, Mainz.

Jaenicke, R., C. Junge and H. J. Kanter, 1971: Messungen der

Aerosolgrössenverteilung über dem Atlantik. *"Meteor"-Forsch.-Erg. Reihe*, **B7**, 1–54.

Junge, C., 1952: Die Konstitution des atmosphärischen Aerosols. *Ann. Meteor.*, **5**, Beiheft.

——, 1963: *Air Chemistry and Radioactivity*. Pergamon Press, 113–123.

Medalia, A. I., and L. W. Richards, 1972: Tinting strength of carbon black. *J. Colloid Interface Sci.*, **40**, 233–252.

Mie, G. 1908: Beitrag zur Optik trüber Medien. *Ann. Phys.*, **25**, 377–445.

Orr, C., F. K. Hurd, W. P. Hendrix and C. Junge, 1958: The behavior of condensation nuclei under changing humidities. *J. Meteor.*, **15**, 240–242.

Volz, F. E., 1972: Infrared refractive index of atmospheric aerosol substances. *Appl. Opt.*, **11**, 755–759.

——, 1973: Infrared optical constants of ammonium sulfate, Sahara dust, volcanic pumice and flyash. *Appl. Opt.*, **12**, 564–568.

Wall, E., 1942: Zur Physik der Wasserdampfkondensation an Kernen. *Z. Angew. Math. Phys.*, **59**, 106–125.

Winkler, P., and C. Junge, 1972: The growth of atmospheric aerosol particles as a function of relative humidity. Part I: Method and measurements at different locations. *J. Rech. Atmos.*, Henri Dessens memorial volume, 617–638.

Yamamoto, G., and M. Tanaka, 1972: Increase of global albedo due to air pollution. *J. Atmos. Sci.*, **29**, 1405–1412.