The optical constants of atmospheric aerosol particles in the 7.5–12 μm spectral region

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ABSTRACT

Thin films of atmospheric aerosol particles which have been collected by an automatic jet impactor are used to obtain the real and imaginary part of the mean complex refractive index of aerosol particles of various types in the 7.5–12 μm spectral region. A dispersion analysis of the measured ir-spectra of absorption of the particle films is performed. By applying the Lorentz theory of continuous dielectrics to the particle films a mathematical relation between the optical constants of the particle films and those of the aerosol particles themselves is obtained. The resulting complex refractive indices are applied to Mie computations to indicate the importance of scattering and absorbing aerosol particles to the radiative heat transfer in the longwave window of the atmosphere.

1. Introduction

Electromagnetic radiation is scattered and absorbed in the atmosphere by aerosol particles. These processes are successfully described by the Mie-theory as a realistic approximation (e.g. van de Hulst, 1957, Bullrich, 1964). Scattering and absorption is determined by the real and imaginary part of the complex refractive index of the aerosol particles as the most important parameters besides the size of the particles, their size distribution, and their concentration in the air. Despite a lot of information on the last three parameters there still is a need of the optical constants of aerosol particles to establish a satisfactory model of the radiative transfer in the atmosphere.

Considering the effect of aerosol particles on the longwave radiative transfer in the atmosphere the $7.5 \leqslant \lambda \leqslant 12$ μm spectral region is of special interest for the following reasons: (1) According to the temperature of the earth–atmosphere-system the maximum of the terrestrial radiation lies between 10 and 12 μm. (2) For $7.5 \leqslant \lambda \leqslant 12$ μm there is a transparent part in the absorption spectrum of atmospheric gases, i.e. the optical thickness of the atmospheric aerosol can be comparable to that of the atmospheric gases. (3) Absorption measurements on the residues of precipitation (Volz, 1972, 1973), as well as on thin films of impactor-deposited particles (Fischer, 1975), and on aerosol particles of in situ state (Grassl, 1973*a*) revealed absorption bands between 8 and 10 μm, which are due to sulfate and silicate compounds contained in the aerosol particles of continental origin.

Therefore, the longwave radiative transfer of the atmosphere is effected by the radiative properties of atmospheric aerosol particles mostly in the $7.5 \leqslant \lambda \leqslant 12$ μm window region: (1) Atmospheric aerosol emits radiation according to its temperature and its absorbing properties causing cooling rates being noticeable in atmospheres of relatively low water vapor content (Grassl, 1973*b*, 1974). (2) In the spectral region considered there is dispersion of the refractive index in case of absorption bands of aerosol particles, i.e. for certain spectral intervals the real part of the refractive index is relatively large. Since scattering is increased by rising of the real part of the refractive index there are spectral regions where aerosol particles may participate in the radiative transfer by scattering despite the small particle size–wavelength proportion.

In this paper it will be shown how the infrared spectra of the mass absorption index of

thin films of aerosol particles previously reported (Fischer, 1975) can be used to get the real and imaginary part of the mean complex refractive index of the aerosol particles themselves in the 7.5–12 μm wavelength interval. At first, a derivation of the macroscopic optical properties of a thin film in relation to the optical properties of the particles contained in the film will be outlined. Secondly, the real part of the complex refractive index will be gained by the dispersion analysis of the spectrum of the measured absorption index of the aerosol films. Thus real and imaginary part of the mean complex refractive index of collectives of aerosol particles are obtained. Additionally some results of Mie calculations using the obtained data on the complex refractive indices will be demonstrated.

2. The optical properties of aerosol films and aerosol particles

The derivation of the relationship between the measurable optical constants of an aerosol film and those of the aerosol particles, which are forming the aerosol film, is based on the elementary theory of continous dielectrics according to Lorentz. Due to their natural size distribution in the atmosphere (Junge, 1963) and to the properties of the jet impactor, that was used for collecting aerosol particles, more than 95% (by number) of the deposited aerosol particles are of radii $r < 0.4$ μm, i.e. much smaller than the wavelengths ($\lambda \geqslant 7.5$ μm) of the irradiation considered. Therefore, the aerosol film having the macroscopic complex refractive index $\hat{n}$ may be treated as a homogeneous medium which consists of small particles of the complex refractive index $\hat{n}$ and the enclosures of air in between. Furthermore, the following model is chosen: the particles forming the film are assumed to be of ellipsoid shape—all particles being of the same volume, but oriented and shaped due to statistically equal likelihood within the aerosol film. This model is as general as possible without making the mathematical problem impractical.

In particular: an external electromagnetic field induces a macroscopic dielectric polarization P_m in the aerosol film which in turn is proportional to the macroscopic electromagnetic field E in the aerosol film:

$$P_m = \frac{\hat{\varepsilon} - 1}{4\pi} E \tag{1}$$

$\hat{\varepsilon}$ = macroscopic, i.e. measurable, complex dielectric constant.

Furthermore, the macroscopic dielectric polarization P_m is given by the number m of the averaged microscopic dipole moments $\langle p \rangle$ of the ellipsoids, s.c. aerosol particles, contained in the whole volume V_g of the aerosol film:

$$P_m = \frac{m \cdot \langle p \rangle}{V_g} = \frac{\bar{\varrho} \cdot \langle p \rangle}{\varrho V} = \frac{\bar{\varrho}}{\varrho} \langle P \rangle \tag{2}$$

where $\bar{\varrho}$ is the density of the aerosol film and ϱ is the density of the particles, V is the volume of a particle, $\langle P \rangle$ is the averaged dielectric polarization of the ellipsoids. Thus, for the macroscopic dielectric constant:

$$\hat{\varepsilon} = 1 + 4\pi \langle P \rangle \frac{\bar{\varrho}}{E\varrho} \tag{3}$$

The average dielectric polarization $\langle P \rangle$ of the ellipsoids is obtained from elementary considerations: (1) The polarization P of an individual ellipsoid embedded in the aerosol film of the dielectric constant $\hat{\varepsilon}$ is given by the Lorentz local field (Kittel, 1973). (2) This polarization P is averaged with respect to all possible orientations of the ellipsoid relative to the direction of the macroscopic field. Isotropy in the aerosol film may be assumed. (3) This mean polarization is further averaged with respect to all shapes of the ellipsoids. Thereby statistically equal distribution of the depolarization factors is assumed between 0 and 4 π (in c.g.s. units), i.e. there is no special shape of the ellipsoids preferably occurring. The resulting relation between the macroscopic dielectric constant $\hat{\varepsilon}$ and the dielectric constant of the particles $\hat{\varepsilon}$ is given by

$$\hat{\varepsilon} = 1 + \frac{2\bar{\varrho}[(\hat{\varepsilon}/\hat{\varepsilon} - 1) \ln \hat{\varepsilon} - 1]}{\varrho - \bar{\varrho}\{1 - (2/\hat{\varepsilon} - 1)[(\hat{\varepsilon}/\hat{\varepsilon} - 1) \ln \hat{\varepsilon} - 1]\}} \tag{4}$$

The real and imaginary part of the refractive index are related to the dielectric constant by the equations:

$$\bar{n} = \left\{\tfrac{1}{2}\left(|\hat{\varepsilon}| + \bar{\varepsilon}_1\right)\right\}^{1/2}$$

$$\bar{k} = \left\{\tfrac{1}{2}\left(|\hat{\varepsilon}| - \bar{\varepsilon}_1\right)\right\}^{1/2} \tag{5}$$

Figs. 1–7. Results of the dispersion analysis and of the calculations of the complex refractive index of aerosol particles in the 7.5–12 μm wavelength interval. Left abscissa: absorption index, right abscissa: real part of the refractive index. *, absorption index $\bar{k}(\lambda)$ measured on the aerosol film; Δ, absorption index $\bar{k}'(\lambda)$ obtained by the best fit; □, absorption index $k(\lambda)$ of the aerosol particles; ○, real part $n(\lambda)$ of the refractive index of the aerosol particles. The dispersion parameters of the best fit are quoted for each sample (cf. eq. 6).

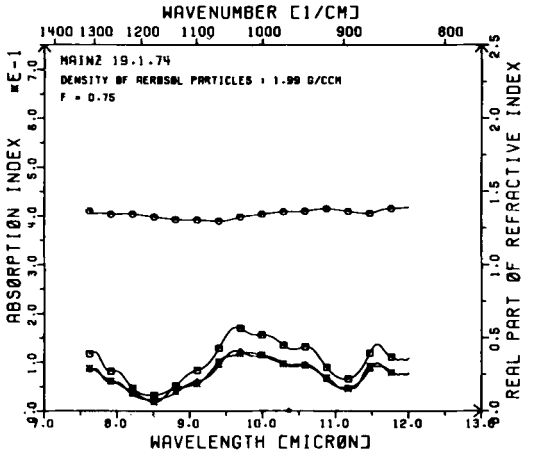

Fig. 1. Aerosol particles collected in urban air at Mainz, Germany.

	$\lambda_j[\mu m]$	$s_j^{\frac{1}{2}}[10^{13}\ sec^{-1}]$	$\gamma_j[10^{13}\ sec^{-1}]$
1	8.016	1.54	1.09
2	7.512	2.31	0.16
3	7.700	1.92	0.94
4	8.996	1.17	1.14
5	11.569	1.41	0.63
6	9.599	2.44	1.39
7	10.105	2.00	1.34
8	10.653	1.51	0.92
9	12.040	0.63	0.21
10	11.800	0.43	0.28
11	11.915	0.43	0.22

$\varepsilon_\infty = 1.42$

where $\bar{\varepsilon}_1$ is the real part of the complex dielectric constant. Thus by inversion of eq. (4) and with eqs. (5) the real and imaginary part of the mean complex refractive index of aerosol particles is obtained from the macroscopic optical constants $\bar{n}$ and $\bar{k}$ of the aerosol film.

3. Dispersion analysis

The absorption index $\bar{k}$ of the aerosol films is determined by the measurement of the mass absorption index $\bar{k}/\bar{\varrho}$ (Fischer, 1975). The quotient $\bar{\varrho}/\varrho$ presents the sum of the volumes of all the particles of a sample divided by the total volume of the aerosol film. $\bar{\varrho}/\varrho$ is set 0.75 which is an average value of related problems (Manegold, 1955). In order to solve eq. (4) for the optical constants of the particles the remaining unknown quantity $\bar{n}$, the real part of the macroscopic refractive index, is calculated via a dispersion analysis using the measured $\bar{k}(\lambda)$-spectrum of the macroscopic absorption index.

It has been shown that in the spectral region of lattice vibrations the dielectric constant is represented by a sum over classical oscillators (Spitzer & Kleinmann, 1961):

$$\hat{\varepsilon}(\omega) = \varepsilon_\infty + \sum_{j=1}^{N} \frac{s_j}{\omega_j^2 - \omega^2 + i\gamma_j\omega} \qquad (6)$$

where ω_j is the resonance frequency of the oscillator j, γ_j its damping constant, and s_j is a factor involving the electric charge, the mass, the spatial concentration, and the strength of the oscillator j. ε_∞ is the contribution of vibra-

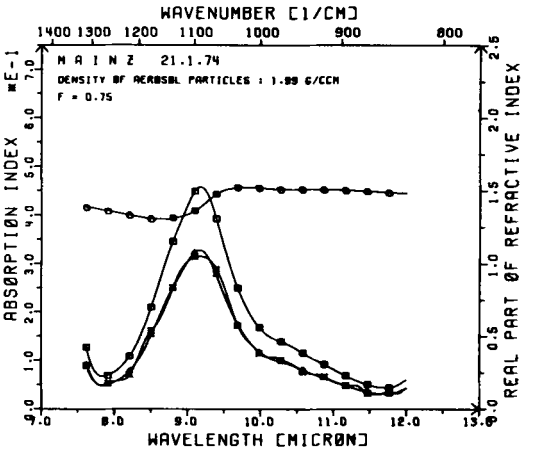

Fig. 2. Aerosol particles collected in urban air at Mainz, Germany.

	$\lambda_j[\mu m]$	$s_j^{\frac{1}{2}}[10^{13}\ sec^{-1}]$	$\gamma_j[10^{13}\ sec^{-1}]$
1	7.546	2.05	0.76
2	8.770	3.28	2.14
3	9.234	4.91	1.84
4	10.912	1.22	1.67
5	9.582	2.25	2.37
6	10.401	1.38	1.40
7	12.009	0.55	0.36

$\varepsilon_\infty = 1.56$

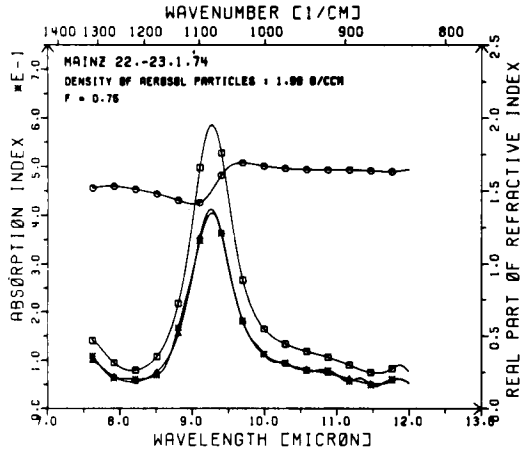

Fig. 3. Aerosol particles collected in urban air at Mainz, Germany.

	$\lambda_j[\mu m]$	$s_j^{\frac{1}{2}}[10^{13}\ sec^{-1}]$	$\gamma_j[10^{13}\ sec^{-1}]$
1	7.578	3.56	2.29
2	10.934	1.94	2.38
3	9.205	4.04	1.41
4	10.243	2.09	2.67
5	9.371	3.91	1.27
6	11.892	0.88	0.55

$\varepsilon_\infty = 1.80$

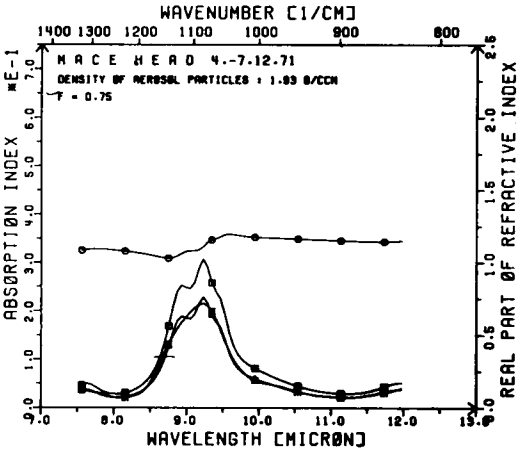

Fig. 4. Aerosol particles collected on the western coast of Ireland.

	$\lambda_j[\mu m]$	$s_j^{\frac{1}{2}}[10^{13}\ sec^{-1}]$	$\gamma_j[10^{13}\ sec^{-1}]$
1	7.622	1.48	1.45
2	8.911	2.49	1.06
3	9.883	2.15	2.84
4	9.246	2.12	0.70
5	9.474	1.41	0.59
6	11.973	1.07	1.12

$\varepsilon_\infty = 1.12$

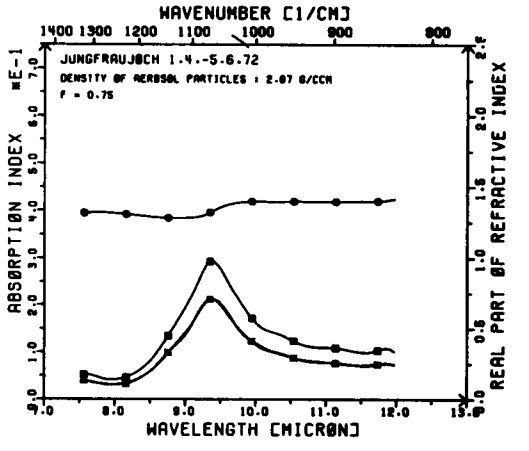

Fig. 5. Aerosol particles collected on Jungfraujoch (3 573 m), Switzerland.

	$\lambda_j[\mu m]$	$s_j^{\frac{1}{2}}[10^{13}\ sec^{-1}]$	$\gamma_j[10^{13}\ sec^{-1}]$
1	7.629	1.67	1.99
2	8.861	2.18	2.09
3	9.327	2.97	1.42
4	11.797	1.93	1.59
5	10.406	2.16	1.91
6	9.660	2.31	1.50
7	11.059	0.74	0.65

$\varepsilon_\infty = 1.45$

tions of high frequencies to the dielectric constant. All these dispersion parameters can be determined by a dispersion analysis. A computer program has been set up that—proceeding from tentative values of the dispersion parameters and for a fixed number of oscillators—calculates the dielectric constant $\hat{\varepsilon}'(\omega)$ and the corresponding absorption index $\hat{k}'(\omega)$ according to eqs. (6) and (5) in the 7.5–12 μm interval. The calculated spectrum of the absorption index $\hat{k}'(\omega)$ is then compared to the measured spectrum of the absorption index $\hat{k}(\omega)$. If the deviation between the two spectra is greater than the mean experimental error of the absorption measurements, the procedure is continued by varying the dispersion parameters, until the experimental data are fitted by the calculated ones within the experimental error. In this case the real part $\hat{n}(\omega)$ of the complex refractive index is computed with the dispersion parameters of the best fit. If the described procedure results in no fit to the experimental spectrum of the absorption index, the number of the oscillators is varied. This method yields the correct values of the optical constants

within the spectral interval considered, even if oscillators of resonance frequencies outside the interval are chosen for fitting the data inside the interval (Verleur, 1968).

Thus the optical constants of aerosol particles can be obtained by:

1. producing a film of aerosol particles by an impactor,
2. measuring the absorption index $\bar{k}(\lambda)$ of the aerosol film,
3. making a dispersion analysis to determine the real part $\bar{n}(\lambda)$ of the complex refractive index, and
4. applying the Lorentz theory of dielectric continua to the aerosol film.

4. Samples of aerosol particles

Uniform films of aerosol substance are produced by deposition of aerosol particles in an automatic jet impactor: the collecting substratum is continuously moved to and fro below slit-like nozzles which atmospheric air is jetting through. The samples investigated were taken at urban and remote sites.

(*a*) Urban air particles: Mainz (145 m), Germany.

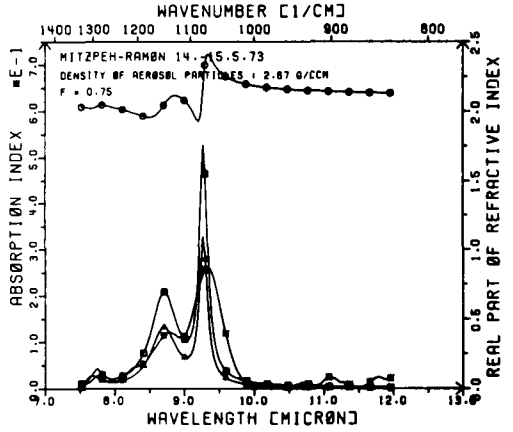

Fig. 6. Aerosol particles collected during a dust storm on High Negev, Israel.

	$\lambda_j[\mu m]$	$s_j^{\frac{1}{2}}[10^{13}\ sec^{-1}]$	$\gamma_j[10^{13}\ sec^{-1}]$
1	7.739	0.99	0.55
2	8.700	3.30	1.13
3	9.271	2.59	2.15

$\varepsilon_\infty = 3.08$

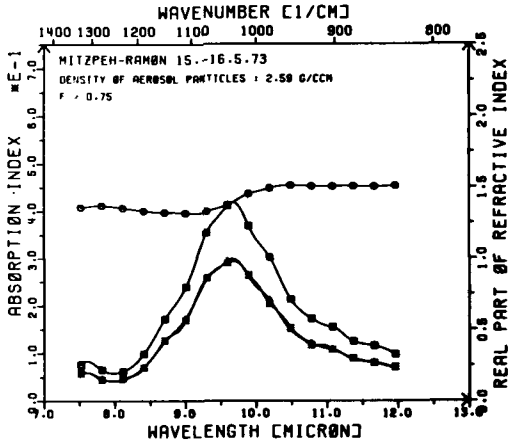

Fig. 7. Aerosol particles collected on High Negev, Israel.

	$\lambda_j[\mu m]$	$s_j^{\frac{1}{2}}[10^{13}\ sec^{-1}]$	$\gamma_j[10^{13}\ sec^{-1}]$
1	7.766	1.56	2.49
2	8.861	2.70	1.48
3	11.182	1.66	2.68
4	10.030	2.25	1.41
5	9.308	2.65	1.16
6	10.723	0.53	0.51
7	10.414	0.46	0.35
8	9.668	1.44	0.74

$\varepsilon_\infty = 1.63$

(*b*) Particles collected at remote sites: Mace Head (30 m), Rep. of Ireland, a rocky promontory on the Atlantic coast; Jungfraujoch (3 573 m), Switzerland; Mitzpeh Ramon (950 m), Israel, a remote site on High Negev.

The mean bulk densities of the aerosol particles collected are summarized in the following table (Thudium, 1975):

Sample	Density [g/cm³]
Mainz 19–23.1.74	1.99
Mitzpeh Ramon 14–15.5.73	2.67
Mitzpeh Ramon 15–24.5.73	2.59
Mace Head 4–7.12.71	1.93
Jungfraujoch 1.4–5.6.72	2.87

5. Results

Results of the computations according to the considerations outlined before are presented in Figs. 1–7. In each diagram the spectrum of the absorption index $\bar{k}(\lambda)$ was plotted that was obtained by the transmission measurements on

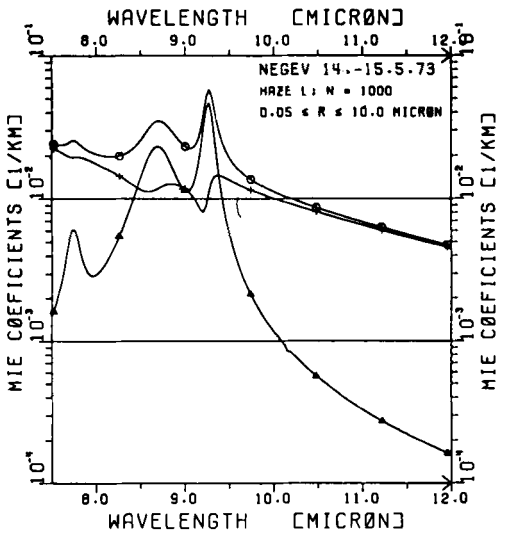

Fig. 8. Coefficients of extinction (○), scattering (+), and absorption (Δ) according to Mie computations. Aerosol particles of quartz-like origin (Fig. 6). Size distribution: Haze L. Total number of particles $N = 1\,000$. Radius interval: $0.05\ \mu m \leqslant r \leqslant 10\ \mu m$.

the aerosol film. There also is a plot of the absorption index $k'(\lambda)$ yielded by the set of the dispersion parameters belonging to the best fit to the spectrum of the experimental absorption index. The dispersion parameters according to eq. (6) are quoted for each example. Additionally the spectra of the real and imaginary part of the mean complex refractive index of the aerosol particles were drawn which were obtained by application of the Lorentz theory of continuous dielectrics to the films of deposited aerosol substance.

Three examples of urban aerosol particles were selected in order to demonstrate the possible variation of the optical properties of aerosol during a sequence of days (Figs. 1–3). The sample collected on Mace Head (Fig. 4) is a mixture of continental and marine aerosol. The sample taken on Jungfraujoch (Fig. 5) consists of particles of mineral and sulfate compounds. Aerosol particles of Mitzpeh Ramon 14–15.5.73 collected during a dust storm belong to a quartz-like substance. The sample of Mitzpeh Ramon 15–16.5.73 was taken at the beginning of a period of meteorological subsidence and is of sulfate and mineral origin.

Details concerning the ir-spectrum of the mass absorption index of the aerosol samples mentioned have been discussed elsewhere (Fischer, 1975).

The broad absorption band between 8.5 μm and 10 μm is a common feature of all continental aerosol particles investigated so far. According to the dispersion analysis this absorption band is realized by two or three oscillators of different strength. The spectra of the real part of the refractive indices exhibit the characteristic properties of spectral dispersion: decrease with decreasing wavelength within the halfwidth of the absorption bands and increase everywhere else. For the types of aerosol particles investigated the real part of the refractive index ranges between 1.0 and 1.7, in some cases above 2.

The limits of the dispersion analysis should be mentioned, too. Obviously there is no use of performing a dispersion analysis of aselective absorption spectra as those of e.g. marine aerosol in the 8–12 μm range (Fischer, 1975). These absorption spectra can be realized by an indefinite number of oscillators. Furthermore, the Lorentz oscillator model seems to fail on certain conditions defined by the relations of solid state physics. It will be noted that some of the experimental absorption bands are broader than is realized by the Lorentz oscillator model (Fig. 6). This broadening has been observed in the case of particles that are much smaller than the wavelength of irradiation, and may be caused by strong, frequency-dependent damping of the oscillators (Genzel & Martin, 1972). However, dense packing of the particles on the substratum has apparently no broadening effect to the absorption bands (Steyer, Day & Huffman, 1974). That implies that the properties of absorption of the aerosol particles are not altered if they are transferred from the airborne to the deposited state. Therefore, the optical constants obtained by the described method are applicable to aerosol particles suspended in the air (Fischer & Grassl, 1975).

6. Applications

Some examples concerning scattering and absorption of collectives of aerosol particles which are suspended in the air are demonstrated. Scattering and absorption of radiation of initial intensity I_0 traversing a distance x is described by the Lambert-formula

$$I = I_0 \exp\{-(\sigma_{sc} + \sigma_{ab})x\} \quad (7)$$

where σ_{sc}, σ_{ab} stands for the coefficient of scattering, resp. of absorption. In the case of aerosol particles of a continuous size distribution σ_{sc}, resp. σ_{ab} is given by:

$$\sigma_{sc/ab} = \int_{r_2}^{r_1} dr \cdot Q_{sc/ab}(r, \lambda, \hat{n}) \cdot \pi \cdot r^2 \cdot n(r) \quad (8)$$

r_1 and r_2 mark the upper and the lower limit of the interval of radii of the size distribution considered. $Q_{sc/ab}(r, \lambda, \hat{n})$ are the factors of efficiency of scattering, resp. absorption; they are functions of the size and the complex refractive index of the particles as well as of the wavelength and are obtained by Mie-calculations. $n(r)$ is the number of particles of radius r per unit volume and per unit increment dr of radius r. The models of the differential particle concentrations $n(r)$ proposed by Deirmendjian (1969) and called Haze L, Haze H, and Haze M were applied to the examples presented here.

The spectra of the coefficients of extinction, scattering, and absorption are shown in Figs. 8–11. In general, in the infrared scattering is not negligibly small compared to absorption, for the ratio between the coefficients of scattering and absorption is dependent on: (1) the dispersion of the refractive index, (2) the size distribution of the particles considered.

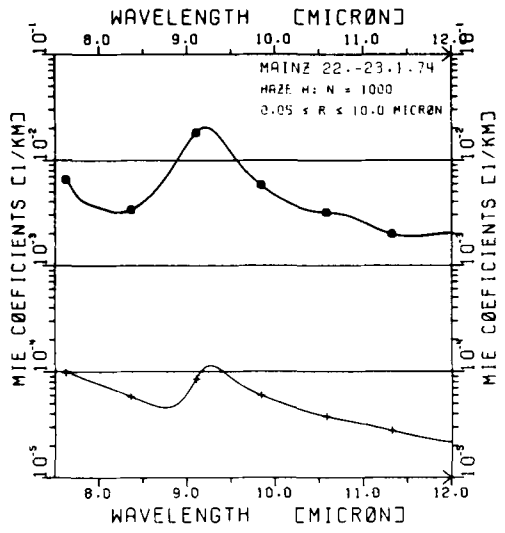

Fig. 9. Coefficients of extinction (○), scattering (+), and absorption (Δ) according to Mie computations. Aerosol particles of urban air (Fig. 3). Size distribution: Haze H. Total number of particles $N = 1\,000$. Radius interval: 0.05 μm $\leqslant r \leqslant$ 10 μm.

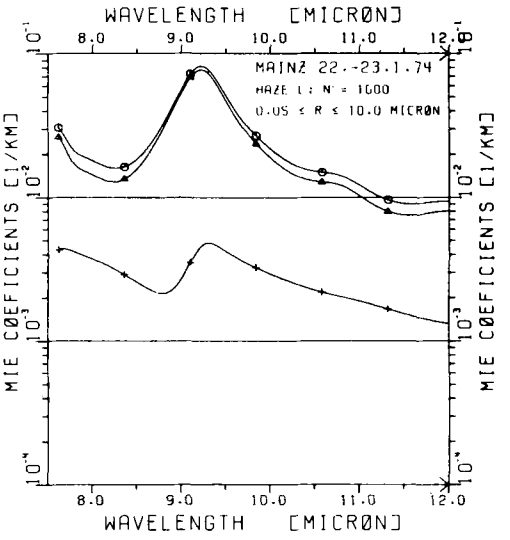

Fig. 10. Coefficients of extinction (○), scattering (+), and absorption (Δ) according to Mie computations. Aerosol particles of urban air (Fig. 3). Size distribution: Haze L. Total number of particles $N = 1\,000$. Radius interval: 0.05 μm $\leqslant r \leqslant$ 10 μm.

(1) In the vicinity of an absorption band the real part of the refractive index at first decreases with increasing wavelength. Thus scattering is reduced. In the spectral range of 'anomalous' dispersion within the absorption band the real part of the complex refractive index is increased so that scattering is increased also. In spectral regions of minor absorption outside a band extinction of radiation may be caused by scattering solely (Fig. 8).

(2) Concerning the size distributions of aerosol particles the scattering coefficient is influenced by the relatively high content of those particles for which the scattering coefficient is proportional to r^4 in the spectral range considered here. In Figs. 9–11 the spectra of the coefficients of extinction, scattering, and absorption were plotted according to the three size distributions mentioned above. The size distributions Haze L, M, and H differ by the radius r_m of maximum differential particle concentration, which is positioned at 0.07 μm, 0.05 μm, and 0.1 μm, resp. The radii r_c, above which the slope of the differential particle concentration is steeper than r^{-4}, are 0.63 μm,

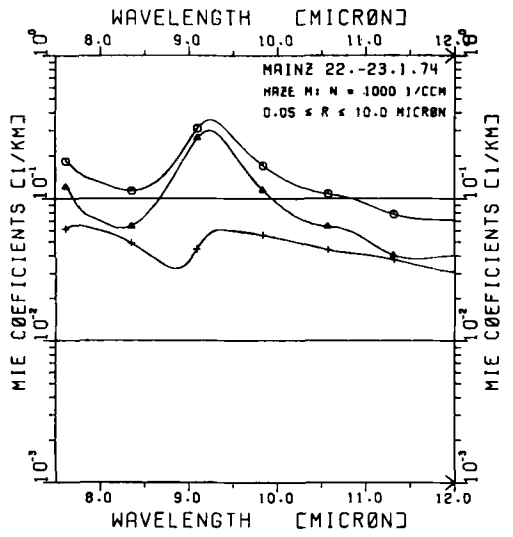

Fig. 11. Coefficients of extinction (○), scattering (+), and absorption (Δ) according to Mie computations. Aerosol particles of urban air (Fig. 3). Size distribution: Haze M. Total number of particles $N = 1\,000$. Radius interval: $0.05\ \mu m \leqslant r \leqslant 10\ \mu m$.

1.25 μm, 0.3 μm, resp., for the three size distributions Haze L, M, and H. The Haze H distribution yields only minor scattering compared to absorption (Fig. 9). Whereas for the Haze L distribution the scattering coefficient amounts to 10% of the extinction coefficient (Fig. 10), in the case of the Haze M distribution the scattering coefficient has increased to about 50% of the extinction coefficient (Fig. 11). Additional computations have shown the crucial importance of the particles of the 1–10 μm radius interval, even if there are relatively few in the natural size distribution of atmospheric aerosol particles.

7. Remarks

In the $8 \leqslant \lambda \leqslant 10$ μm spectral region atmospheric aerosol particles of continental origin posses absorption bands which can be assigned to sulfates and silicates as their natural compounds. As these absorption bands coincide with the spectral range of the atmospheric longwave transparency a marked influence of continental aerosol on the radiation budget of the atmosphere can be expected, especially in cases of enlarged aerosol concentrations caused by temperature inversions and in atmospheres of low water vapor content (Grassl, 1973*b*). Furthermore, because of spectral dispersion of the refractive index the properties of scattering of aerosol particles are dependent on the wavelength so that generally scattering by aerosol particles is not negligibly small compared to absorption by aerosol in the longwave atmospheric window.

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ОПТИЧЕСКИЕ КОНСТАНТЫ АТМОСФЕРНОГО АЭРОЗОЛЯ

Для получения действительной и мнимой частей комплексного показателя преломления частиц аэрозолей различного типа в области спектра 7,5–12 *мкм* использовались тонкие пленки аэрозольных частиц, собранных автоматическим струйным коллектором. Выполнен дисперсионный анализ измеренных ИК-спектров поглощения пленок частиц. При использовании теории Лоренца сплошных диэлектриков для описания пленок получено математическое соотношение между оптическими константами пленок и соответствующими величинами для самих аэрозольных частиц. Полученные значения комплексного показателя преломления используются в расчетах по теории Ми для выяснения роли рассеивающих и поглощающих частиц аэрозоля в радиационном переносе тепла в длинноволновом окне атмосферы.