

REFINED OPTICAL CHARACTERISTICS OF AEROSOLS IN THE GROUND LAYER OF THE ATMOSPHERE IN MEXICO

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Model calculations of the characteristics of light scattering in the ground layer of the atmosphere in Mexico have been carried out taking account of uncertainties in the data of electrostatic and photoelectric counters of particles. The results obtained in this study demonstrate an important contribution of variations in a coarse aerosol fraction to the variability of light scattering characteristics.

Model calculations of the optical characteristics of aerosol in the ground layer of the atmosphere in Mexico were done in Ref. 1 for dry and wet seasons on the basis of the available data on the microstructure parameters of aerosol particles. In spite of the fact that microphysical measurements were performed in the ecologically cleanest region of Mexico, the calculated values of the aerosol optical characteristics seemed to be essentially underestimated. The errors caused by uncertainty of the complex refractive index may not result in significant underestimating of the values of scattering and extinction coefficients. (As the refractive index varied from $n = 1.55$ to $n = 1.65$, the values of the coefficients increased by 10–20% for the particle size distributions under consideration.)

The errors in determining the aerosol particle size distributions can be caused by two reasons: 1) underestimating of the particle size measured by an electrostatic spectrometer (TSI-2000) and 2) incorrect extrapolation of the particle size distribution function for $r \geq 0.4 \mu\text{m}$.

In spring of 1992 we carried out integrated measurements of aerosol characteristics that allowed us to refine the chemical composition of aerosol particles and hence to set more justified complex refractive index for calculations as well as to obtain the particle size distribution function for $r \geq 0.2 \mu\text{m}$ by means of the AZ-5 photoelectric counter.

The following estimates of the chemical composition of aerosol in the ground layer of the atmosphere in Mexico were obtained from the data of neutron-activation, mass-spectrometer, and IR-spectral analyses of filter samples: mineral components primarily of soil origin made up $12\text{--}40 \mu\text{g}/\text{m}^3$, chloride salts amounted to $2.0\text{--}7.0 \mu\text{g}/\text{m}^3$, nitrates constituted $2.0\text{--}6.0 \mu\text{g}/\text{m}^3$, sulfates and sulfuric acid made up $5.0\text{--}35 \mu\text{g}/\text{m}^3$, soot amounted to $2.0\text{--}12.0 \mu\text{g}/\text{m}^3$, organic substances constituted $0.5\text{--}2.5 \mu\text{g}/\text{m}^3$, and the content of water in the dry aerosol matter was about $3.0 \mu\text{g}/\text{m}^3$. Total measured mass concentration varied from 32 to $106 \mu\text{g}/\text{m}^3$.

Such chemical composition at low relative humidity ($f_{\text{H}_2\text{O}} < 40\%$) corresponds to the real part of the complex refractive index $n = 1.6\text{--}1.63$, or approximately to $1.5\text{--}1.58$ in dry season and to $1.42\text{--}1.5$ in wet season in Mexico.

When measuring by the TSI-2000 electrostatic spectrometer, the aerodynamical particle size was determined. In general, it is not equal to its geometrical size² governing the optical characteristics of particles (scattering, absorption, and extinction cross sections, scattering phase function, and degree

of linear polarization). The experimentally measured values $\Delta N_i(r)$ in the range of particle radii $\Delta r_I = 0.003\text{--}0.281 \mu\text{m}$ (range I) shift toward larger radii $\Delta r_{II} = 0.0038\text{--}0.375 \mu\text{m}$ (range II). In Ref. 1 the size distribution function for large particles was obtained by extrapolation on the basis of the power-law function $f(r) = dN/dr = C r^{-4}$.

The data of measurements by photoelectric counter in 1992 allowed us to reveal some peculiarities of the particle size distribution function in the range $\Delta r = 0.2\text{--}10 \mu\text{m}$ for Mexico. Its salient features are the deviations from the power-law function in the ranges $\Delta r_{III} = 0.2\text{--}0.7 \mu\text{m}$ and $\Delta r_{IV} = 1\text{--}10 \mu\text{m}$, i.e., the increase of the particle number density. The first is caused by the processes of photochemical generation of aerosol matter immediately in the polluted urban air, namely, by formation of aerosol particles in the process of combustion of petrol in automobile engines. The second is caused primarily by raising dust above various surfaces. The first mode obviously exhibits the diurnal variations, and the second mode variations are primarily determined by the weather conditions (Fig. 1).

Thus the data on the disperse composition of aerosol particles allow us to estimate the contribution of the uncertainty in determining the particle size distribution function in the atmosphere in Mexico using different aerosol devices and techniques for extrapolation of the measured incomplete particle size spectrum to the aerosol optical characteristics.

The variations of the extinction and scattering cross sections of individual spherical particles in the range of particle size $0.003\text{--}0.375 \mu\text{m}$ for the complex refractive index $m = (1.47 - i \cdot 0.003)$ produce modest increase of these parameters. However, calculations for polydispersed systems (in ranges I and II of particle size) show a significant (approximately threefold) increase of the extinction and scattering cross sections for the shifted minimum and maximum particle radii from the ascertain data of the electrostatic spectrometer. It should be noted that the relative volume occupied by the scattering particles and calculated by the formula

$$V_{\text{aer}} = \frac{4}{3} \pi \int_{r_{\text{min}}}^{r_{\text{max}}} dN/dr r^3 dr$$

also increases. Thus in wet season for range I it was $34.57 \mu\text{m}^3/\text{cm}^3$, and for range II it was $81.84 \mu\text{m}^3/\text{cm}^3$. In dry season it was 48.3 and $114.4 \mu\text{m}^3/\text{cm}^3$, respectively.

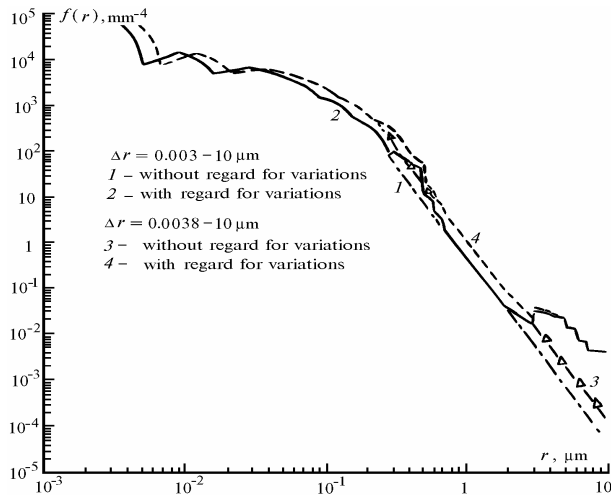


FIG. 1. Model particle size distribution functions for dry season in Mexico taking into account the variations of $f(r)$ in the ranges of particle size $\Delta r_{III} = 0.2 - 0.7 \mu\text{m}$ and $\Delta r_{IV} = 1 - 10 \mu\text{m}$.

For dry season, with the particle size distribution function extrapolated by the power-law function, the extinction coefficient was $30.83 \cdot 10^{-3} \text{ km}^{-1}$ for $\lambda = 0.55 \mu\text{m}$ in the range of particle size $0.003 - 10 \mu\text{m}$. It corresponds to the meteorological visibility range $S_m = 127 \text{ km}$, i.e., it is essentially underestimated. Taking into account the shift of particle size ($0.0038 - 10 \mu\text{m}$), it is equal to $80.11 \cdot 10^{-3} \text{ km}^{-1}$ or $S_m = 49 \text{ km}$, that is, more realistic for the conditions of the clean urban atmosphere. Meteorological visibility range was calculated by the formula $S_m = 3.9 / K_{\text{ext}}$ for $\lambda = 0.55 \mu\text{m}$.

Much larger variations of aerosol optical characteristics are found to occur with regard for the variations of the particle size spectrum in the ranges of particle size $\Delta r_{III} = 0.2 - 0.7 \mu\text{m}$ and $\Delta r_{IV} = 1.0 - 10.0 \mu\text{m}$.

For dry season the results of calculation of the extinction coefficients K_{ext} are presented in Table I for the ranges of particle size $0.003 - 10$ and $0.0038 - 10 \mu\text{m}$ with and without regard for the variations of the particle number density in the ranges of particle size Δr_{III} and Δr_{IV} .

TABLE I.

Range, $\lambda, \mu\text{m}$	$K_{\text{ext}} \cdot 10^{-3}, \text{ km}^{-1}$			
	Range of particle size, μm			
	0.003-10	0.0038-10	0.003-10	0.0038-10
	Without regard for variations	Without regard for variations	With regard for variations	With regard for variations
0.35	40.28	94.79	72.67	128.46
0.45	35.47	89.12	72.03	127.33
0.55	30.83	80.11	69.87	121.34
0.65	26.89	70.60	65.95	112.16
0.75	23.67	62.22	61.19	102.26
0.85	20.96	55.30	56.52	93.31
1.0	17.97	46.88	50.57	81.53

In the study of the effect of the particle size distribution function on the angular scattering characteristics (scattering phase function and degree of linear polarization), it should be noted that the shift of the minimum scattering particle size from $\Delta r = 0.003 \mu\text{m}$ to $\Delta r = 0.0038 \mu\text{m}$ for any wavelengths in the range

$\lambda = 0.35 - 1 \mu\text{m}$ has practically no effect on the shape of the scattering phase function.

Transformation (by way of additions) of the distribution function in the ranges Δr_{III} and Δr_{IV} increases the portion of the forward scattered light, i.e., results in higher degree of forward-peaking of the scattering phase function. The shape of the scattering phase function depends to a greater extent on the inner structure of scattering particles (their inhomogeneity and nonsphericity).¹

The transformations of the spectrum connected with the shift of the minimum scattering particle radius from $r = 0.003 \mu\text{m}$ to $r = 0.0038 \mu\text{m}$ produce more essential distortions in the shape of the degree of linear polarization. These distortions are observed at the angles both smaller and larger than 90° .

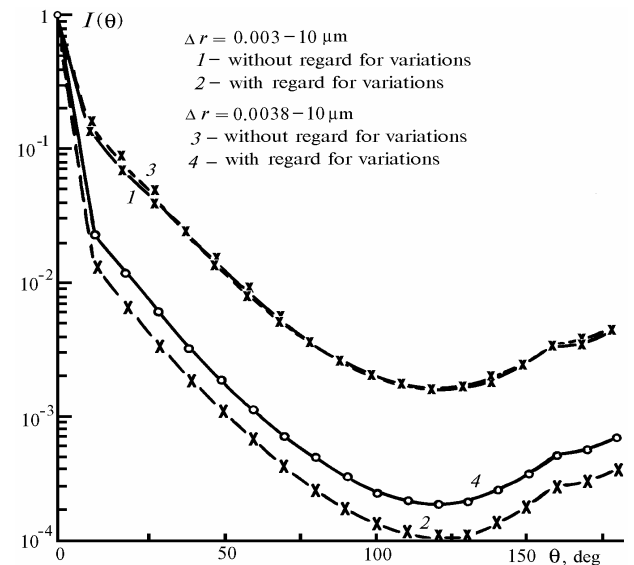


FIG. 2. Scattering phase functions $I(\theta)$ at $\lambda = 0.55 \mu\text{m}$ for dry season in Mexico with and without regard for the variations of $f(r)$ in the ranges of particle size $\Delta r_{III} = 0.2 - 0.7 \mu\text{m}$ and $\Delta r_{IV} = 1 - 10 \mu\text{m}$.

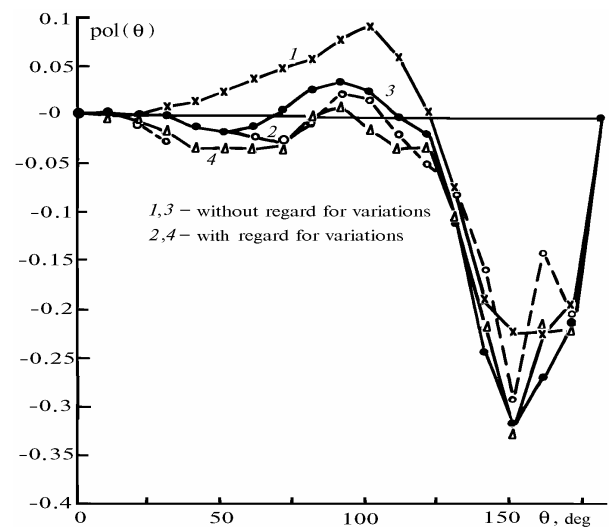


FIG. 3. Degree of linear polarization $pol(\theta)$ at $\lambda = 0.55 \mu\text{m}$ for dry season in Mexico with and without regard for the variations of $f(r)$ in the ranges of particle size $\Delta r_{III} = 0.2 - 0.7 \mu\text{m}$ and $\Delta r_{IV} = 1 - 10 \mu\text{m}$.

For dry season the results of calculations of the scattering phase functions and degree of linear polarization are shown in Figs. 2 and 3 for $\lambda = 0.55 \mu\text{m}$ in the ranges of particle size $0.003\text{--}10 \mu\text{m}$ (curves 1 and 2) and $0.0038\text{--}10 \mu\text{m}$ (curves 3 and 4).

The similar pattern is observed in calculations of aerosol optical characteristics of the ground layer of the atmosphere in wet season in Mexico.

The variations of the angular scattering characteristics make it possible to monitor the variations of the atmospheric aerosol microstructure from the optical data.

The results obtained confirm quite obvious conclusion that in model calculations of the light

scattering characteristics one should take into account the particle size distribution function more accurately, especially for coarse fraction of particles. Particularly, one should take into account the instrumental errors of different aerosol devices as well as the strong variability of the coarse fraction of atmospheric aerosols.

REFERENCES

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