REMOTE METHOD FOR DETERMINING THE PATH-AVERAGED INDEX OF REFRACTION OF AEROSOL PARTICLES

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The statistical relations between the index of refraction of aerosol particles, the coefficient of asymmetry of aerosol scattering of light, the coefficient of asymmetry of the backscattering phase function, and Angstrom's parameter, which characterizes the spectral behavior of the scattering coefficients (optical thickness), were investigated by analyzing 110 *models of the atmospheric aerosol.*

The dependences established were approximated by analytical relations that make it possible to estimate to within 1.3–3.8% *the path-averaged value of the index of refraction of aerosol particles from the polydispersion characteristics of the aerosol.*

The obtained estimates of the index of refraction can be used to invert the measured optical characteristics of the aerosol into the microphysical characteristics.

In the last few years, the development of computational methods and means has made it possible to determine the size spectrum and index of refraction of aerosol particles from data on the optical properties of the aerosol.[1]

Iteration procedures which involve simultaneously the search for the optimal value of the index of refraction and the determination of the histogram of particle sizes continue to remain, however, quite unwieldy and require large amounts of computer time. This restricts the possibility and range of practical applications of these methods and programs for processing large arrays of optical measurements.

The solution of the problem is significantly simplified mathematically and the required computer time is reduced by many factors if the effective index of refraction of optically active particles is known *a priori* (at least approximately).

As is correctly pointed out, for example, in Ref. 2, however, aerosol structures are characterized by exceptional diversity and variability. For this reason, in many cases, it is impossible to choose a sufficiently reliable *a priori* value of the index of refraction of the particles.

Analysis of the solutions of direct problems of aerosol optics shows that the spectral behavior of the extinction coefficient (optical thickness) and the form (coefficient of asymmetry) of the light-scattering phase function of aerosol are sharp functions of the index of refraction of the particles. Therefore, if it were possible to determine, at least approximately, the index of refraction of aerosol particles from data on the spectral behavior of the scattering coefficient and the coefficient of asymmetry of the scattering phase function, then it would be much easier to solve the inverse problems of aerosol optics.

The problem addressed in this paper is to express by means of very simple semiempirical relations the dependence of the index of refraction on the indicated optical characteristics of aerosol.

Angstrom's index ω in the formula $\beta(\lambda) = \alpha\lambda^{-\omega}$ is most often used to characterize the spectral behavior of the aerosol extinction coefficient $\beta(\lambda)$.

This index is closely related with the particle-size spectrum. In the case of an inverse power-law particle-size distribution

$$dN(r)/d\log r \sim r^{-\nu^*}$$

for a quite wide spectrum of particle sizes $\omega = \nu^* - 2$.

We shall characterize the form (asymmetry) of the aerosol scattering phase function $\gamma(\theta)$ by the asymmetry coefficient

$$\Gamma = \int_0^{\pi/2} \gamma(\theta)\,\sin\theta\,d\theta \Big/ \int_{\pi/2}^{\pi} \gamma(\theta)\,\sin\theta\,d\theta.$$

The coefficient Γ depends primarily on the index of refraction m of the particles, but this dependence is not single-valued. As the index of refraction changes (for example, owing to flooding of the particles during development of the condensation processes) different types of redistribution of particles over sizes cam occur, and therefore the asymmetry coefficient Γ and the index ω will vary differently. In particular, as the number of large particles increases, as a rule, the index ω decreases and the asymmetry coefficient Γ increases, but this happens differently in different cases.

A conclusion of this type, purely of a qualitative character, is drawn in the publications of

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many authors, but usually no quantitative relations were presented.

Based on the stochastic character of the processes and the conditions of aerosol formation, the relations sought should also be of statistical character. For this reason, it was natural to try to determine these relations by making a statistical analysis of the existing optical models of aerosol.

Data on thousands of scattering phase functions of polydisperse aerosols for different combinations of real and imaginary parts of the index of refraction $m = n - \kappa i$, for different types of particle-size distribution functions and the parameters of such functions, combinations of minimum and maximum particle sizes, etc. have now been published.

However the spectral behavior of the scattering coefficients (optical thicknesses), based on which the index ω can be determined, is by no means presented in all cases. The asymmetry coefficient Γ is given even less often.

We selected from an array of scattering phase functions with known parameters m, ω, and Γ the particular phase functions that best conform to the actually observed functions. We focused first on the adequacy of the conditions of formation of the backscattering part of the phase function at scattering angles in the range $\theta = 90$–$180°$ (backscattering phase function). This part of the phase function is most sensitive to a change in the index of refraction (at small angles the scattering phase function is virtually independent of the index of refraction).

In Ref. 3 and the works of other authors it is shown that in the formation of the real scattering phase functions the microparticle fraction ($r < 0.1$ μm) makes a large contribution to the backscattering part of the scattering phase function. When there are too few microparticles, a situation that is characteristic for unimodal bell-shaped log-normal and γ distributions, an unrealistically deep minimum forms in the backscattering part and this results in overestimation of the asymmetry coefficient Γ and deformation of the spectral behavior of the scattering coefficients toward lower values of the Angstrom index ω.

Based on this, we eliminated from the analysis aerosol models with a deficiency of microparticles and we studied only models with moderate values of the asymmetry coefficient and Angstrom's index. The scattering phase functions calculated by S.V. Dyshlevskii[4] and O.M. Korostina (in an agreement on scientific collaboration) for continental and marine aerosols with a mean-weighted particle-size spectrum,[5] formed a significant part of the set of scattering phase functions employed for analysis.

In all we employed for analysis 110 scattering phase functions, calculated by different authors for the region of the spectrum from 0.347 to 1.06 μm, indices of refraction m from 1.33 to 1.65–0.005i, parameters ω from 0.37 to 1.8, and asymmetry coefficients Γ from 5.4 to 16.5. For values of $k \le 0.015$ it was assumed that $m = n$. Cases with $k > 0.015$ were not studied.

Analysis established that for values of $\omega \le 1.6$ the relation between the parameters Γ, ω, and m can be approximated comparatively by the expression

$$\Gamma \simeq a/(m - 0.4)^2(2 + \omega^2)$$

or

$$m^{\mathrm{I}} \simeq 0.4 + \sqrt{a/\Gamma(2 + \omega^2)}, \qquad (1)$$

where $a \simeq 33$.

To estimate the accuracy of the formula (1) we compared the values of m^{I} obtained with their help with the starting values of m (used in the Mie calculations).

The calculations showed that the average (δ) and rms (σ^{I}) errors in the determination of the index of refraction $\overline{\delta^{\mathrm{I}}} = N^{-1}\sum_1 \delta_1^{\mathrm{I}} = 1.92\%$ and

$$\sigma^{\mathrm{I}} = \sqrt{(N-1)^{-1}\sum_1 (\delta_1^{\mathrm{I}} - \delta^{\mathrm{I}})^2} = 3.75\%$$

for $\omega \le 1.6$ and $N = 91$.

For $\omega > 1.6$ it is recommended that the formula (1) not be used, since strongly underestimated values of the index of refraction can result.

In addition, analysis of selected scattering phase functions established that the ratio $\eta = \gamma(120° - 30°)/\gamma(120° + 30°)$, characterizing the asymmetry of the backscattering phase function, is closely related with the index of refraction m. The larger the index of refraction m the larger the asymmetry coefficient η is, and this relation is very stable and is virtually independent of the particle-size distribution, i.e., it is independent of the Angstrom index ω.

Statistical analysis showed that on the average this dependence is approximated well for $\omega > 0$ by the expression

$$m^{\mathrm{II}} = 1.3 + \eta^2\sqrt{\omega}/3\pi \qquad (2)$$

The average ($\overline{\delta^{\mathrm{II}}}$) and rms ($\sigma^{\mathrm{II}}$) errors in the estimates of m based on this formula are $\overline{\delta^{\mathrm{II}}} = 0.8\%$, $\sigma^{\mathrm{II}} = 3.12\%$ for $N = 110$.

Thus if for the aerosol model under study the values of Γ and ω are known, then m can be estimated using the formula (1); if η and ω are known, then m is estimated using the formula (2). In most cases the errors in the estimates made with the formulas (1) and (2) have different signs. Thus if it is possible to calculate the values m^{I} and m^{II} using the formulas (1) and (2), then for the estimate of the index of refraction m it is best to use the value

$$m = \frac{m^{\mathrm{I}} + m^{\mathrm{II}}}{2}. \qquad (3)$$

The rms error in the estimate of m in this case drops to $\sigma = 1.33\%$, i.e., is decreases by more than a factor of two.

The following, however, should be kept in mind.

All these estimates refer only to situations when the characteristics m, Γ, ω, and η (for both marine and continental aerosols) have moderate values, the scattering phase functions (in the classification given by O.D. Barteneva) are of the gently sloping type, and the particle-size spectrum has a form close to the weighed-mean form.[5] The possibility of using the formulas (1)—(3) to estimate the index of refraction of aerosol particles of other types, differing sharply from the types indicated above, was not studied.

When estimating the errors in the determination of the index m from data obtained by single measurements of the characteristics Γ, ω, and η the error in the measurements of these characteristics must also be taken into account in particular, the formula (2) could turn out to be ineffective owing to the large errors in measurements of the coefficient η.

In conclusion it is my duty to point out that this paper was largely made possible thanks to the calculations of scattering phase functions performed by O.M. Korostina and S.V. Dyshlevskii for polydispersed systems.

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