On the Atmospheric Variability of the Spectral Behavior of the Aerosol Extinction Coefficient

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Introduction

The peculiarities of the radiation budget of the space-atmosphere-earth surface system are determined by the specific character of the processes of transformation of the incident solar radiation and the outgoing infrared (IR) radiation from the underlying surface. To properly take into account the noted processes, adequate data are necessary on the aerosol extinction, scattering, and absorption coefficients corresponding to different climatic zones and states of the optical weather.

The main advantages of the semi-empiric optical models, which are created on the basis of the long-term statistically provided measurements of the spectral transparency of the atmosphere (Kabanov et al. 1988; Krekov and Rakhimov 1982), lie in the adequate representation of actual atmospheric optical events. The disadvantage is the fact that such models are created for a small number of geographical regions. Also, such models describe only the total aerosol extinction without separating absorption and scattering, so they are applicable only in atmospheric transparency windows. Although the so-called "microphysical imitation models" of the aerosol lightscattering parameters provide the possibility of overcoming the disadvantages of the empirical approach (Krekov and Rakhimov 1986; Rakhimov and Panchenko 1999), they are burdened by the imperfection of the initial data on the aerosol microstructure. One of the goals of this paper is to realize a new approach to the creation of the aerosol model that combines the advantages of empirical and theoretical methods.

Methodological Aspects

In this paper, we analyze the results of simulation and the experimental data on the spectral aerosol extinction coefficient on horizontal near-ground paths. The following statistical parameters of corresponding ensembles of theoretical and experimental data are compared: the mean values of the coefficients $\beta(\lambda_k)$, their rms errors $\sigma_{\beta}(\lambda_k)$, as well as the expansions in terms of the orthonormalized system of eigenvectors $\phi_i(\lambda_k)$ of the autocorrelation matrix $B_{\beta\beta}(\lambda_k,\lambda_i)$. The data were obtained in Crimea in 1981 and in Tomsk in 1995. Intensive forest fires in the Siberian region were observed in the fall of 1997. The spectral transparence of the atmosphere was measured in 23 separate spectral intervals in the atmospheric transparency windows in the wavelength range 0.4 μ m to 12 μ m on the horizontal paths. The path length was 1 km in Tomsk and 7.5 km over sea gulfs in Crimea. The duration of separate measurement cycles was from two weeks to two months. During a day, we obtained 6 to 12 mean spectra of the aerosol extinction coefficient calculated from the data on the atmospheric transparency taking into account the molecular absorption of water vapor and trace gases.

The approach used to form the numerical data developed in this paper is based on the imitation of the statistical variety of microstructural characteristics of atmospheric haze (Rakhimov and Panchenko 1999). Variations of the particle size spectrum were simulated based on the hypothesis that the disperse composition of atmospheric haze is the result of interaction of different aerosol sources, each of them affecting a certain size range (see Figure 1). So the aerosol particle size distribution function was simulated as a superposition of lognormal modes (Krekov and Rakhimov 1982, 1986; Rakhimov and Panchenko 1999):



 $f(\mathbf{r}) = \frac{dN}{dr} = \mathbf{A}^{(v)} \mathbf{r}^{-v} \sum_{i=1}^{k} \mathbf{M}_{i}^{(v)} \exp\left\{-\mathbf{b}_{i} \left[\ln\left(\mathbf{r} / \mathbf{r}_{mi}^{(v)}\right)\right]^{2}\right\}$

Figure 1. Some examples of the simulated and measured (Willeke and Whitby 1975) size spectra.

Contrary to Rakhimov and Panchenko (1999), who analyzed the optical properties of atmospheric haze at one wavelength, we now need data on the refractive index of aerosol particles $m(\lambda) = n(\lambda) = i\kappa(\lambda)$ in wide wavelength range. Spectral dependencies of $n(\lambda)$ and $\kappa(\lambda)$ for some chemical compounds (Zuev and Krekov 1986) are shown in Figure 2.

Seventeen types of spectral dependencies $m(\lambda)$ were used for simulating the optical constants taking into account the real chemical composition of atmospheric haze. Optical constants of coarse fraction were simulated using $m(\lambda)$ characteristic of different minerals (external mixing). The data on $m(\lambda)$ for chemical complexes such as a mixture of sulfur acid, water, ammonium sulfate, etc. were used for



Figure 2. Spectral dependencies of $n(\lambda)$ and $\kappa(\lambda)$ for some chemical compounds (Zuev and Krekov 1986).

simulating the fine fraction. Weight proportion between the components were simulated by a random generator with normal distribution around the mean value.

Results

Figures 3 through 5 present the calculated spectral dependencies of $\beta(\lambda_k)$, $\sigma_\beta(\lambda_k)$, and $\phi_i(\lambda_k)$ in comparison with the experimental data obtained in the Black Sea coastal zone, West Siberia, and under conditions of intensive but distant forest fires.

The agreement between calculative and experimental data is reached by selection of the mean values and rms errors of the simulated microstructural parameters, such as modal radius and mode width.

The mean volume distribution functions of aerosol particles are shown in Figure 6. Except for the summer of 1995, only two modes are seen in all distributions V(r), although the hypothesis on some independence of variations of the parameters of four fractions was used when determining the specific realizations.

The dotted lines in Figure 6 show the results of the model approximation of the mean spectra. It is seen that more than a triple increase of the total cross-section of aerosols from secondary sources is observed as relative humidity increases, while the total cross section of aerosol from primary sources increases only by 40%.



Figure 3. Statistical characteristics of the spectral aerosol extinction coefficients obtained near Tomsk in spring and summer 1995 (fine-dispersed haze).



Figure 4. Statistical characteristics of the spectral aerosol extinction coefficients obtained near Tomsk in October 1997 (far forest fire).



Figure 5. Statistical characteristics of the spectral aerosol extinction coefficients obtained on the Black Sea coast in 1981 in conditions of enhanced relative humidity (RH > 70%).

Conclusion

The analysis of the data obtained shows that:

- the content of giant particles (obviously, of organic origin) significantly increases in the atmosphere of West Siberia in summer in comparison with spring and fall
- in conditions of forest fires, not only the content of fine particles increases even far away from the fire center (at 300 km to 450 km), but also the content of the particles of 10 μm to 20 μm. Evidently, it is related to the intensification of the vertical stream of the coarse fraction at the increase of the convective component over big areas of forest fire.

The analysis allowed us to reveal the hierarchy of the processes determining the variability of the optical characteristics and to assess the role of regular and random factors in the mechanism of formation of the statistical variety of the states of atmospheric haze.

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Figure 6. Mean aerosol particle volume distributions obtained from experimental optical data.

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