MODELING OF OPTICAL CHARACTERISTICS OF NEAR–GROUND ATMOSPHERIC AEROSOL IN THE 0.3–15 μm WAVELENGTH RANGE.
III. RESULTS OF MODELING

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A procedure for constructing a model of optical characteristics of the near–ground atmospheric aerosol has been described. The values of the parameters have been presented, and the model results are discussed.

Following principles outlined in Ref. 1 and model consideration of near–ground aerosol composition and structure given in Ref. 2, we have identified three basic aerosol fractions that differ in origin, composition, and the whole of physical and chemical properties. The particle modal radii were estimated to be $r_{0i} \approx 0.03$, 0.3, and 1 μm at the atmospheric humidity $f_0 \approx 50\%$ for fractions 1, 2, and 3, respectively. In addition to these basic fractions typically present in the near–ground aerosols, also considered are the specific aerosol forms such as finely dispersed particles composed of organic matter (with modal size $r_0 \approx 0.03$ μm), water–droplet aerosols, coarsely dispersed industrial aerosols, and smoke. Concentration of these minor aerosol fractions, as well as the particle size distribution of two last aerosol types, depend essentially on local conditions.

We have calculated the optical characteristics of this aerosol types. In Fig. 1, the spectral behavior of coefficients of aerosol extinction and absorption, $\omega^{(i)}(\lambda, f) = \alpha^{(i)}(\lambda, f)/a^{(i)}_{\rm ext}$ ($\lambda = 0.5$ μm, $f = 50\%$), is shown for different values of relative humidity of air and for three basic aerosol fractions. As to the other fractions enumerated above, the dependence of optical properties on air humidity is readily considered for water droplets. Corresponding curves are shown in Fig. 2.

For organic aerosols contributing to transformation of the atmospheric aerosol properties under humidity variations, accurate quantitative estimates, that are at all rigorous, are lacking. We can do no more than mention various related hypotheses typically semiqualitative and speculative.[3] If no interaction of this fraction with other aerosol fractions is suggested (which is doubtful), its contribution to the formation of aerosol optical properties is normally small,[4] and furnishes an opportunity to consider this contribution as a small correction for aerosol optical characteristics, independent of humidity; corresponding data are considered below. Industrial aerosols, as was pointed out in Ref. 2, are difficult to take correctly into account due to their diversity and hence deserve special consideration. A possible contribution of soot particles also calls for separate study[5] due to their specific structure and properties.

Natural atmospheric aerosols represent a mixture of all fractions weighted by the fractional concentration. At the first stage of analysis, these weighting functions were evaluated by comparing calculated curves of spectral behavior of aerosol extinction coefficients to experimental results.[6] As a starting approximation, an assumption that at $f \approx 50\%$, atmospheric aerosol particles are approximately described by the Junge size distribution with $\upsilon = 3.6$ (which is the case in the first approximation), was used. Under varying humidity, this distribution transforms accordingly. For each spectrum being considered, we determined the correction factors accounting for the contribution of the given aerosol fraction to the aerosol extinction coefficient due to deviation of its concentration from the initial model. As preliminary analysis has shown, in most cases aerosol particle size spectra so "reconstructed" differ from the measured ones (in the range of particle size $r \in$ 0.2–10 μm, whose minimum and maximum radii are limited by the capabilities of optical microscopy) by no more than 20–30%, which is within the measurement error.

This allows us to proceed to the next stage of our analysis connected with an effort to establish the dependence of the particular aerosol fractional concentration on atmospheric conditions. With the amount of data available, we cannot establish exact regression dependence. (Whereas in the study of the dependence of aerosol extinction on the air humidity we were able to process over 200 spectra,[7] in the present case, due to extremely cumbersome procedure of simultaneous analysis and processing of experimental optical and microstructural data and calculated results, we had to confine ourselves to only 30 cases.) Thus, based on the semiqualitative relationships so established, we have made an attempt to compare the obtained results with the experimental spectra chosen at random from the total array of experimental data available.

Table I presents results of such comparison. Specifically, under normal atmospheric conditions ($S_m = 20$ km, $f = 50\%$), the aerosol extinction coefficients for three basic aerosol fractions are $a^{(1)}_{\rm ext} = 0.06509$, $a^{(2)}_{\rm ext} = 0.08196$, and $a^{(3)}_{\rm ext} = 0.04832$ km^{-1}, respectively, and the correction factors given in Table I allow one to consider the size spectrum transformation for individual aerosol fractions in the atmosphere as functions of the meteorological visibility range S_m and relative air humidity f (actually, the coefficients $\gamma_i(S_m, f)$ also consider different types of the complex refractive index m dependence on air humidity at $\lambda = 0.5$ μm for individual aerosol fractions).

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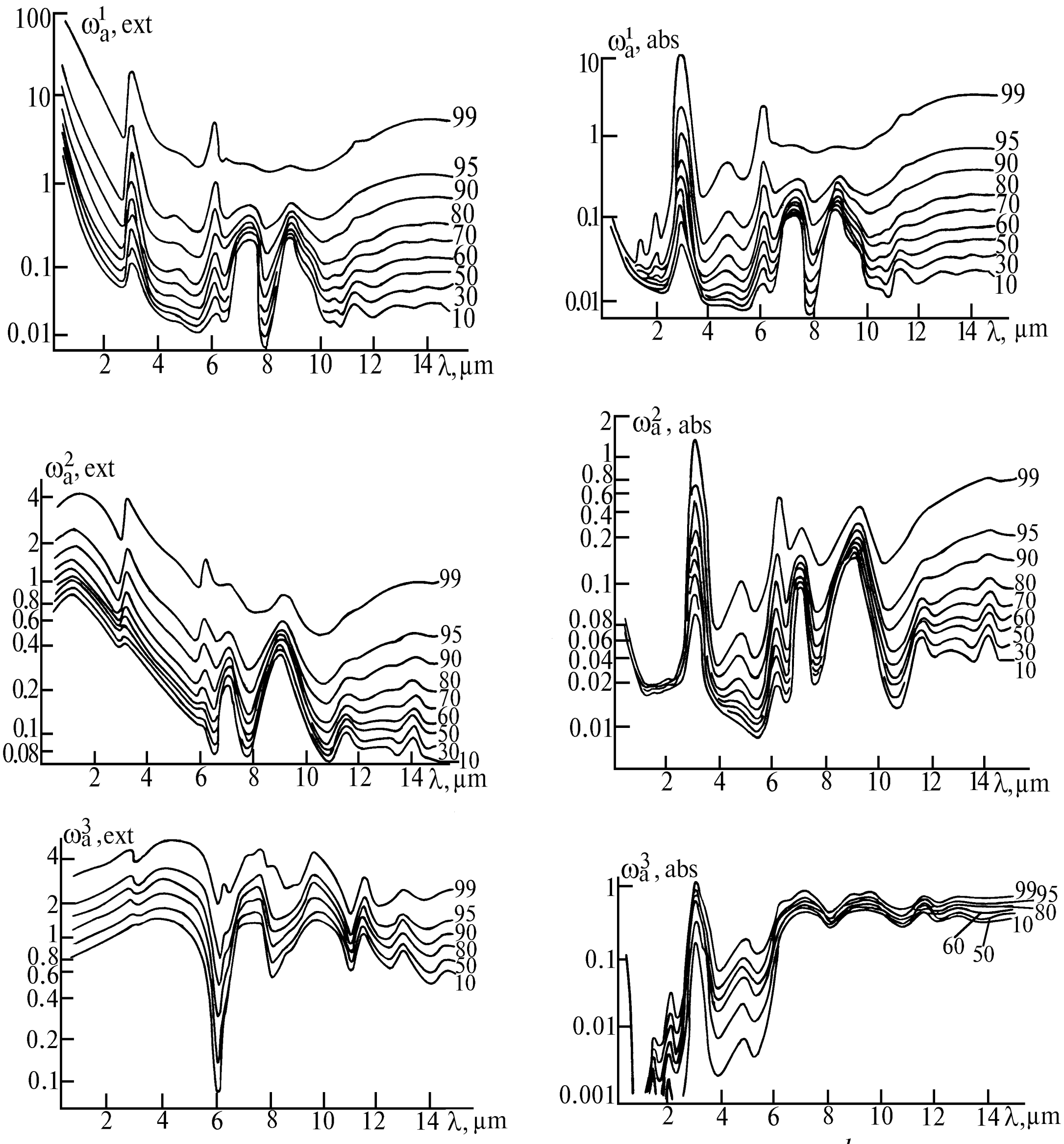
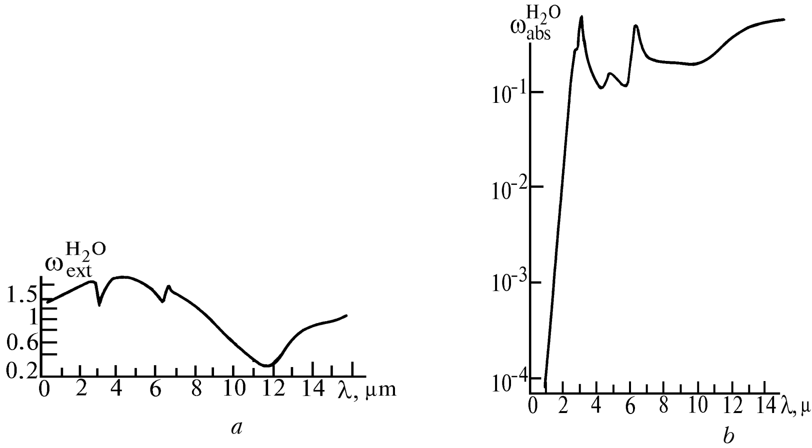


a *b*

FIG. 1. Spectral behavior of the aerosol extinction (a) and absorption (b) coefficients for different values of air humidity.



a *b*

FIG. 2. Spectral behavior of the extinction (a) and absorption (b) coefficients for water droplets.

TABLE I. Weighting functions $\gamma_i(S_m, f)$ *of basic aerosol fractions for different values of meteorological visibility range* S_m (km) *and relative humidity of air* f (%).

f	S_m							
	5	10	15	20	25	30	50	i
10	3.249	2.050	1.573	1.276	1.085	0.961	0.640	1
	4.332	2.343	1.656	1.276	1.044	0.890	0.556	2
	8.045	3.347	1.893	1.276	0.924	0.686	0.318	3
20	3.121	1.961	1.501	1.215	1.032	0.912	0.605	1
	4.161	2.242	1.579	1.215	0.992	0.845	0.526	2
	7.728	3.202	1.805	1.215	0.879	0.651	0.281	3
30	2.973	1.859	1.417	1.144	0.970	0.863	0.566	1
	3.964	2.124	1.491	1.144	0.933	0.794	0.492	2
	7.361	3.035	1.704	1.144	0.826	0.607	0.281	3
40	2.822	1.755	1.333	1.074	0.909	0.800	0.527	1
	3.763	2.006	1.403	1.074	0.874	0.741	0.487	2
	6.988	2.866	1.603	1.074	0.774	0.572	0.262	3
50	2.658	1.644	1.243	1.000	0.845	0.742	0.487	1
	3.543	1.879	1.309	1.000	0.812	0.687	0.424	2
	6.581	2.684	1.496	1.000	0.719	0.530	0.242	3
60	2.378	1.336	1.082	0.864	0.726	0.639	0.412	1
	3.171	1.527	1.139	0.864	0.698	0.592	0.361	2
	5.889	2.181	1.301	0.864	0.618	0.435	0.205	3
70	2.007	1.196	0.881	0.698	0.583	0.505	0.324	1
	2.677	1.367	0.928	0.698	0.560	0.468	0.282	2
	4.971	1.953	1.060	0.698	0.496	0.361	0.161	3
80	1.632	0.951	0.691	0.543	0.450	0.388	0.246	1
	2.176	1.087	0.727	0.543	0.432	0.359	0.214	2
	4.041	1.552	0.831	0.543	0.383	0.277	0.122	3
90	1.085	0.609	0.432	0.335	0.275	0.234	0.146	1
	1.477	0.696	0.455	0.335	0.264	0.217	0.127	2
	2.686	0.994	0.520	0.335	0.234	0.167	0.073	3
95	0.649	0.350	0.243	0.186	0.151	0.128	0.079	1
	0.862	0.400	0.256	0.186	0.146	0.119	0.068	2
	1.602	0.571	0.292	0.186	0.129	0.091	0.039	3
99	0.166	0.085	0.058	0.044	0.035	0.029	0.018	1
	0.211	0.098	0.061	0.044	0.034	0.027	0.015	2
	0.411	0.139	0.069	0.044	0.030	0.021	0.009	3

Obviously, the fact that the regularities, obtained in the above–indicated manner, describe adequately (to within 20 to 30%) experimental data[6] is by no means indicative of their objectivity and, in some sense, their universality. Such conclusion could be made, if at all, by comparing the obtained results with independent experimental data. But it has been just this comparison that presents the main difficulties.

On the one hand, the literature usually provides individual spectra illustrative in character that makes it impossible to form a data array to conduct a test. On the other hand, as was noted in Ref. 1, aerosol extinction in the IR range was experimentally found as a small difference between at least two small values – measured atmospheric extinction and calculated continuum absorption by H_2O. (In most spectral intervals available for infrared measurements in the real atmosphere, proper allowance must be made for the third contributor to atmospheric extinction – the selective absorption by atmospheric gases, also determined by calculation.)

As a result, estimates of aerosol extinction coefficients have poor accuracy, and, what is more significant, the nature and properties of continuum absorption by water vapor are as yet poorly understood. Consequently, with the abundance of techniques currently used for processing and interpretation of measurement results, their intercomparison is problematic even in principle.

Nevertheless, we were fortunate to perform one such comparison, by a detailed analysis of original experimental data[8] (14 spectra), kindly transferred to the Department of Atmospheric Physics of the Scientific–Research Institute of Physics (SRIP) at the St. Petersburg State University by Dr. Brounshtein during joint work. Whereas Ref. 8 treated H_2O absorption theoretically, by the method of Ref. 9, results of Ref. 6 were processed by empirical technique.[10] Difference between the spectral coefficients of aerosol extinction estimated in Refs. 6 and 8 was great (by a factor of 2 to 4), to say nothing of the negative residual (i.e., independent of the amount of water vapor on a ray path) extinction, very often observed in Ref. 8 and entirely absent in Ref. 6.

Measurements reported in Ref. 6 were performed in Tomsk, and those of Ref. 8 – in Voeikovo Settlement (near Leningrad). Unfortunately, optical measurements of Ref. 8 were not accompanied by simultaneous microphysical investigations providing independent data on the structure and properties of measurable atmospheric aerosols. At the same time, aerosol data of sufficient seasonal coverage were available from repeated measurements, though occasional, performed by the Laboratory of Physics of Aerosols of the SRIP in Voeikovo. Aerosol samples taken in Voeikovo were included in data array that provided a basis for the model of the complex refractive index of aerosol matter of individual fractions. Noteworthy, the measurements exhibited no pronounced peculiarities of aerosol in the examined region in comparison with other regions. It is not surprising in this regard that the processing of a portion of data from Ref. 8, available for us, by method of Ref. 10 yielded the estimated aerosol extinction coefficient values in the IR range that were in complete agreement with the results of Ref. 6. Correspondingly, description of data borrowed from Ref. 8 and processed using the procedure described above gives the results practically identical to those presented above. This conclusion may be disproved in case of processing of the entire array of data of Ref. 8, since the aerosol extinction coefficient values obtained theoretically and experimentally by the method of Ref. 10, differ slightly greater than in case of Ref. 6, by 30–50%, but this slightly increased difference also can be attributed to the change in the range of variation of the humidity of air.

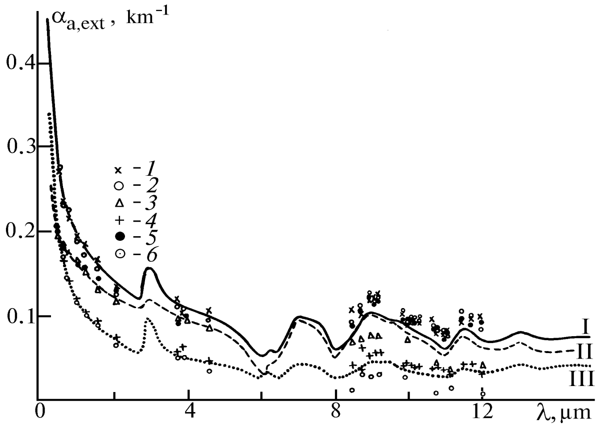


FIG. 3. Calculated (curves) and measured (points) coefficients of aerosol extinction under various atmospheric conditions. Experimental data: 1, 2, 5) Ref. 6; 3, 4, 6) Ref. 8. $S_m = 11$ km, $f = 44\%$ (I); $S_m = 14$ km, $f = 45\%$ (II); $S_m = 19$ km, $f = 81\%$ (III),

Similar comparison was performed based on original data of Ref. 8, by using both published material and spectra available for us. It is natural that difference between experimental data and results of model calculations in this case was much greater. However, we think that these distinctions appear explicable and reasonable. Some results of comparison of model calculations with experimental data from Refs. 6 and 8 are illustrated by Fig. 3, indicating that experimental values of Ref. 6 are typically overestimated relative to calculated ones, while data of Ref. 8 are substantially underestimated (by a factor of 1.5 to 2). We think that these differences are due to the fact that Refs. 6 and 8 employ diverse methods of allowing for IR–radiation absorption by atmospheric water vapor. Method of Ref. 9 supposedly overestimates absorption by H_2O resulting in underestimation of aerosol contribution to infrared radiation absorption, while the empirical method of Ref. 10 underestimates absorption by water vapor thereby overestimating the aerosol contribution.

From the aforesaid, the practical application of the model proposed is as follows: for the given values of meteorological visibility range S_m and relative humidity f choose from Table I the correction coefficients $\gamma_i(S_m, f)$ and, using curves $\omega^{(i)}(\lambda, f)$ of Fig. 1 together with the values $a^{(i)}_{\text{ext},0}$ shown above, calculate the spectral coefficients of aerosol extinction from the formula

$$\alpha_{\text{ext}}(\lambda, S, f) = \underset{i}{\alpha^{(i)}_{\text{ext},0}}\, \omega^{(i)}_{\text{ext}}(\lambda, f)\, \gamma(S_i, f_m) . \qquad (1)$$

The absorption and scattering coefficients are calculated in the same way.

Figure 4 shows a comparison of aerosol extinction coefficients calculated for the model discussed above with the experimental data reported in Ref. 11. In contrast to comparison discussed above, curves of Fig. 4 were calculated with one more fraction of sea–water droplet; their size distribution and concentration were chosen on recommendation of Ref. 4. Naturally, for such a comparison to be made, $\gamma_i(S_m, f)$ values discussed above should be recalculated.

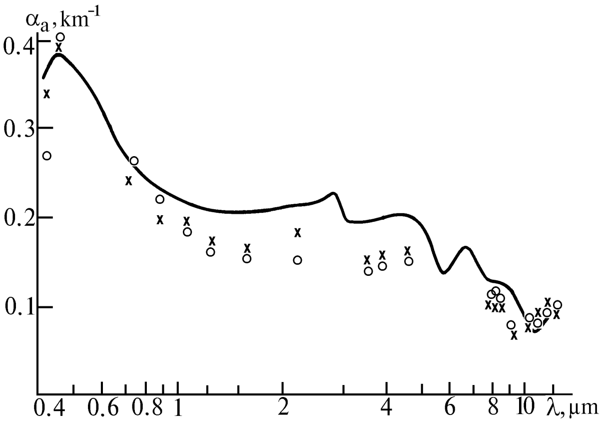


FIG. 4. Comparison between calculated (curves) and measured[11] *(points) values of aerosol extinction coefficient for coastal regions.*

As an example, Fig. 5 shows calculated curves $\alpha_{\text{ext}}(\lambda, S, f)$ in some typical cases. Also shown are the aerosol extinction coefficients for finely–dispersed organic aerosol fraction with concentration chosen to be close to its maximum value reported in Ref. 4.

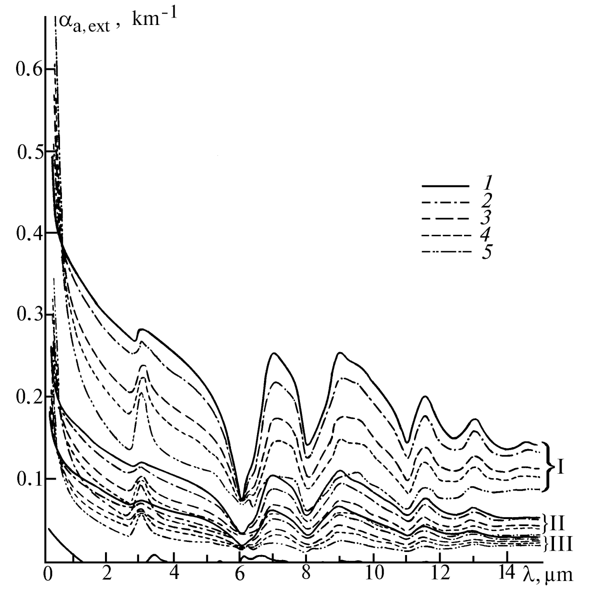


FIG. 5. Calculated aerosol extinction coefficients for S_m = 10 (I), 20 (II), *and* 30 km (III) *and* f = 50 *(1),* 70 *(2),* 80 *(3),* 90 *(4), and* 95% *(5).*

From the analysis of results of comparison between calculated aerosol extinction coefficients and experimental data made in a wide spectral range under various atmospheric conditions, we conclude that the proposed model predicts with confidence, to within 30–50% accuracy, the spectral behavior and absolute values of optical energy characteristics of atmospheric aerosols over the entire 0.3–15 μm wavelength range. The found discrepancy by a factor of 1.5 to 2 seems to be not only permissible, but even advantageous considering the present–day state of the problem. The agreement is much better in the visible range, for λ < 1 μm, where the calculated angular and polarization characteristics for the model described above do not contradict the experimental data.

We also note that the model proposed can be easy extended to the entire atmospheric depth.

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