# INTERPRETATION OF THE MODEL OF SPECTRAL EXTINCTION FOR COASTAL MARINE HAZE

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The results of an interpretation of a single-parameter spectral model of the extinction coefficient of coastal marine haze are presented. The method employed to solve the corresponding inverse problem is examined. The general behavior of the microstructural parameters of coastal haze as a function of the degree of atmospheric turbidity is determined. It is shown that the haze consists of submicron and coarsely dispersed fractions, and the contribution of these fractions to the extinction of visible and IR radiation is estimated.

### 1. INTRODUCTION

The most complete and comprehensive information about the optical characteristics of coastal marine haze and their variability, caused by a number of geophysical factors, are presented in the monograph Ref. 1. In Ref. 2 data on the microstructure and index of refraction of the submicron fraction of haze with particle radii not exceeding 0.8 µm were obtained from the angular dependences of the polarization scattering phase functions of marine coastal haze at the wavelength  $\lambda = 0.546 \ \mu m$  (Ref. 2). The complete collection of energy and polarization characteristics of haze was reconstructed from microstructural data by the method of microphysical extrapolation in the spectral interval  $0.44-0.84 \ \mu m$ . It was noted that extrapolation to the region  $\lambda > 0.84~\mu m$  is unacceptable because the experimental data do not contain enough information about the coarsely dispersed particle fraction ( $r > 0.8-1.0 \mu m$ ).

Together with the angular polarization characteristics of coastal haze, in Ref. 1 considerable attention is devoted to the analysis of the aerosol extinction of optical radiation at wavelengths in the range  $0.48-11.9 \ \mu\text{m}$ . It is natural to expect that interpretation of the spectral dependences of the extinction coefficient of the haze will give additional information about the coarsely dispersed fraction and will provide the prerequisites necessary for extending the spectral range of microphysical extrapolation.

By analogy to Ref. 2, in this paper the object of interpretation are not separate experimental realizations, but rather an average statistical model of the spectral behavior of the aerosol extinction coefficient,<sup>1</sup> written in the form

$$\sigma(\lambda) = k_0(\lambda) + k_1(\lambda) \cdot \sigma(0.55), \tag{1}$$

where  $\sigma(\lambda)$  is the aerosol extinction coefficient at the wavelength  $\lambda$ ,  $\sigma(0.55)$  is the aerosol extinction coef-

ficient at the wavelength  $\lambda = 0.55 \ \mu m$  (this coefficient is an input parameter of the model), and  $k_0(\lambda)$ and  $k_1(\lambda)$  are empirical coefficients. The regression model (1) was constructed by Yu.A. Pkhalagov and V.M. Uzhegov based on analysis of base measurements, performed over a period of many years, of the spectral transmission of the atmosphere along the coastal of the Black Sea and describes the spectral variability of the extinction coefficient of haze as a function of the degree of atmospheric turbidity. This model is based on statistical analysis of about 1000 spectra, obtained over six seasons on three paths at different points of the coast of the Black Sea. The model makes it possible to estimate the aerosol extinction coefficient for a number of wavelengths  $\lambda_1$ from the interval  $0.48-11.9 \ \mu m$  based on its value at the wavelength  $\lambda = 0.55 \ \mu m$ . Analogous statistical models with the same input parameter  $\sigma(0.55)$  were developed previously for the components of the scattering matrix for continental haze<sup>3</sup> and for the polarization scattering phase functions of coastal haze.

In this paper an interpretation of the single-parameter model (1) is given based on the solution of the inverse problem in the spectral interval 0.55–3.9  $\mu$ m in order to estimate the microstructural parameters of haze, whose optical manifestations would be consistent with the model (1). The solution of this problem will make it possible to determine the nature of the transformation of the dispersed composition of the haze accompanying a change in the degree of atmospheric turbidity, and to enlarge, on this basis, the data base for constructing a microstructural model of coastal haze.

# 2. METHOD FOR SOLVING THE INVERSE PROBLEM

The mathematical formulation of the problem of inverting the spectral dependence of the aerosol extinction coefficient is discussed in detail in Ref. 5 and consists of solving the integral equation

$$\int_{0}^{R} \mathcal{K}(\lambda, r) s(r) dr = \sigma(\lambda), \qquad (2)$$

where s(r) is the distribution function, to be determined, of the particles over the sizes of the geometric cross section; r is the radius of the particles;  $K(\lambda, r)$  is the extinction efficiency, which depends on the complex index of refraction  $m - i \cdot \kappa$ ; and, the right side  $\sigma(\lambda)$  is specified by the model (1). The undetermined parameters in Eq. (2) are the index of refraction  $m - i \cdot \kappa$  and the maximum particle size R. Since the questions regarding the method for inverting equations of the form (2) have been studied many times in the literature, we shall give only a brief description of the method employed; details will be discussed as necessary.

A regularizing algorithm, developed based on the method of A.M. Tikhonov and used previously for interpreting angular polarization measurements,<sup> $\delta$ </sup> was used to invert Eq. (2). The vector of the approximate solution s was found by inverting regularized system of linear algebraic equations equivalent to Eq. (2):

$$[K^*K + \alpha D]s_{\alpha} = K^*\sigma, \qquad (3)$$

where  $K^*$  is the matrix conjugate to the matrix K; D is a smoothing matrix; and,  $\alpha$  is a regularization parameter.

TABLE I. Spectral dependence of the real part of the index of refraction m for different visibility ranges  $S_m$ .

	λ, µm	S <sub>m</sub> , km				
		5	10	20	30	50
	0.55	1.38	1.39	1.41	1.43	1.44
	0.69	1.38	1.39	1.41	1.43	1.44
	1.06	1.37	1.39	1.40	1.42	1.43
	1.2	1.37	1.38	1.40	1.415	1.425
	1.6	1.355	1.365	1.38	1.39	1.40
	2.2	1.32	1.32	1.33	1.34	1.34
	3.9	1.375	1.375	1.38	1.385	1.39

**2.1.** Estimate of m and  $\kappa$ . To calculate the kernel  $K(\lambda, r)$  the variability of the spectral behavior of  $m(\lambda)$  and  $\kappa(\lambda)$  for different values of the visibility range  $S_m$  was determined from previous interpretations of the single-parameter model of the polarization scattering phase functions of the haze of interest in the visible range ( $\lambda = 0.55 \ \mu m$ ).<sup>2</sup> In Ref. 2 estimates of m(0.55) and  $\kappa(0.55)$  were obtained, and data on the volume fill factors of the two fractions – "dry matter"  $V_d$  and water  $V_w$  – and their variability for different values of the meteorological visibility range  $S_m$  were obtained. In particular, the quantity m increases

monotonically from 1.38 to 1.44 as  $S_m$  varies from 5 to 50 km. This is connected with the increase in the relative fraction of the "dry matter" from 30% for  $S_m = 5$  km up to 70% for  $S_m = 50$  km. The spectral dependences  $m(\lambda)$  and  $\kappa(\lambda)$  were calculated, using the formula for the specific refraction, based on data on the ratio of the volume fill factors  $V_d$  and  $V_w$  with different visibility range  $S_m$ . The data on the spectral behavior. of the index of refraction of each fraction were taken from Ref. 7. The results of the calculation of  $m(\lambda)$  are presented in Table I. In the spectral range0.55–3.9 µm the imaginary part of the index of refraction  $\kappa(\lambda)$  assumes small values, which do not exceed 0.005 at  $S_m = 5$  km and 0.01 at  $S_m = 50$  km.

**2.2.** Estimate of R. The a priori choice of the upper limit of integration in Eq. (2) was based on a modification of the method proposed in Ref. 5. The lower limit of the maximum particle size  $R^*$  was estimated by an iteration method according to the scheme

$$\begin{pmatrix} R_{n}^{\bullet} \end{pmatrix}^{-1} \int_{0}^{R_{n}^{\bullet}} K(\lambda, r) dr = \sigma(\lambda)/S_{n-1}, \quad n = 1, 2, ...,$$

$$S_{n} = \int_{0}^{R_{n}^{\bullet}} S_{n}(r) dr,$$

$$(4)$$

where  $s_n(r)$  is the distribution reconstructed by inverting Eq. (2) with the upper limit  $R = R_n^*$ ; S is the total geometric cross section of the aerosol particles per unit volume; and, *n* is the iteration number. In this method the initial value  $s_0$  must be known. In the case at hand the initial value  $S_0$  was estimated from the solution of the inverse problem for the polarization phase functions.<sup>2</sup> For example, for the meteorological visibility range 5 km the initial value  $S_0 = 0.37$  km<sup>-1</sup>. Solving Eq. (4) for  $\lambda = 3.9 \ \mu m$  gives  $R_1^* = 1.7 \ \mu m$ . For the distribution  $s_1(r)$ , reconstructed by inverting Eq. (2) with the upper limit  $R = R_1^*$ , the new value is  $S_1 = 0.28 \text{ km}^{-1}$ . The next iteration of Eq. (4) for  $S = S_1$  gives in turn the new estimate  $R_2^* = 1.9 \ \mu m$ . In this approach two iterations are sufficient in practice in order to obtain an estimate of the lower limit of values of  $R^*$  that is difficult to improve in further iterations. This is attributable to the fact that within wide limits variations of the *a priori* value of *R* have virtually no effect on the variability of the total cross section S of the reconstructed microstructural distributions. Thus, for example, when the domain of the function s(r) is extended up to R = 4.3 µm. i.e., it is approximately doubled, the value of S remains practically unchanged and equal to 0.3 km<sup>-1</sup>. This method of estimating  $R^*$  for visibility  $S_m$  in the range 5–50 km gives close results in the range 1.9-2.3 km<sup>-1</sup>.

A different variant of estimating R\* is based on *a* priori knowledge of the polydispersion extinction efficiency  $\overline{K}_m = \sigma_{max} / S$  at the extremum point of the

spectral dependence  $\sigma(\lambda)$ .<sup>5</sup> In this case the equation for estimating  $R^*$  has the form

$$(R^{\bullet})^{-1} \int_{0}^{R} K(\lambda_{\max}, r) dr = \overline{K}_{m} \sigma(\lambda_{\max}) / \sigma_{\max}.$$
 (5)

According to Ref. 5, for a wide class of atmospheric hazes of the type H, L, and M, the range of variation of the values of  $\overline{K}_m$  is estimated to be 2.6–3.3. For the present model (1) a maximum in the spectral function  $\sigma(\lambda)$  is observed only when the atmospheric transmission is high and for  $S_m = 50$  km it is reached at the point  $\lambda = 0.69$  µm. Taking into account the range of variation of the factor  $\overline{K}_m$  an estimate of  $R^*$  based on the formula (5) gives a value in the range 2.2–2.55 µm, which is in good agreement with the previously obtained results.

The family of solutions s(r), reconstructed from the spectral dependence  $\sigma(\lambda)$  (1), corresponding to  $S_m = 5$  km, is shown in Fig. 1 for an increasing sequence of values of R. The initial value of R is equal to 2.05  $\mu$ m. Further correction of *R* toward increasing values was made in a manner so that the total contribution of particles with sizes greater than R to the optical characteristics would be negligibly small. One can see from Fig. 1 that for  $R = 3 \mu m$  the function s(r) drops to zero; this gives a basis for setting the final value  $R = 4.3 \ \mu m$  for the upper limit of the range of dispersion of the haze under study. Analysis of the solutions presented in Fig. 1 shows that increasing Rhas no effect on the character of the distribution of particles with sizes less than 0.9 µm, while the coarsely dispersed fraction "spreads" over an increasingly wide interval with a corresponding decrease in the average level. It is interesting to note that all distributions s(r) with a stable total cross section  $S \simeq 0.28 - 0.3 \text{ km}^{-1}$  correspond optically to the model (1) with approximately the same error (7-8.5%).

**2.3. Choice of the parameter \alpha.** When the regularized solution of Eq. (2) is constructed the value of the regularization parameter a must be consistent with the error in the starting data. However, when the right side of (2) is given by some model of the form (1), there arise difficulties associated with the correct estimate of the error of the model (1). Under these conditions, it is better to use criteria, for choosing a that do not depend explicitly on the errors in the starting data. As methods of this type, selection based on the criterion that the value of the regularization parameter  $\alpha_q$  be quasioptimal<sup>8</sup> as well as based on the principle of minimum discrepancies<sup>8</sup>  $\alpha_{md}$  were studied. In the latter case the value of a was determined from the condition that the functional

$$\rho = \|Ks_{\alpha} - \sigma\| + \|KPs_{\alpha} - \sigma\|,$$

where P is the projection operator on the set of nonnegative functions:

$$Ps_{\alpha}(r) = \begin{cases} s_{\alpha}(r), & s_{\alpha}(r) \ge 0, \\ 0, & s_{\alpha}(r) < 0 \end{cases}$$

be minimum. In the problem under study these criteria give close values of  $\alpha$ , and in addition  $\alpha_{md} < \alpha_q$  and the discrepancy is equal to 5–8%.



FIG. 1. The results of inversion of the spectral model of the aerosol extinction coefficient  $\sigma(\lambda)$  for  $S_{\rm m} = 5$  km with a different choice of the maximum particle size R.

2.4. Correction of the finely dispersed fraction. Because of the approximate character of the solution of the inverse problem the value of the extinction coefficient  $\sigma(\lambda)$ , calculated from the reconstructed microstructure  $s_{\alpha}(r)$  at the wavelength  $\lambda = 0.55 \ \mu m$ , differs from the value  $\sigma(0.55)$  – the input parameter of the optical model (1). As an example, Fig. 2 shows the spectral dependences  $\sigma(\lambda)$ , corresponding to the model (1) for  $S_m = 20$  km (curve 1) and the results of the calculation of  $\sigma = Ks_{\alpha}$  (curve 2), the disagreement between which at the point  $\lambda = 0.55 \ \mu m$  is equal to about 10%. There arises a situation in which the aerosol distribution  $s_{\alpha}(r)$ , reconstructed from the optical model (1) with a given visibility range in reality gives a visibility range different from the model value. For this reason there is only an approximate correspondence between the input parameter of the optical model (1) and its microstructural image. It is thus necessary to correct the solution  $s_{\alpha}(r)$  in order to compensate for the indicated discrepancy in the optical characteristics. It should be noted that the extinction coefficient in the short-wavelength region can be compensated without appreciable distorting other sections of the spectrum only by correcting the finely dispersed aerosol fraction. Eliminating from the total extinction  $\sigma(\lambda)$  the contribution of the extinction  $\sigma_c(\lambda)$ determined by the coarsely dispersed fraction  $(r \ge 1 \ \mu m)$  and having a neutral behavior in the region  $\lambda \leq 1.2 \mu m$ , we obtain the spectral dependence of the extinction coefficient  $\sigma_m(\lambda) = \sigma(\lambda) - \sigma_c(\lambda)$  of the submicron fraction of the haze. From the solution of the inverse problem for  $\sigma_m(\lambda)$  we obtain the corrected size distribution of the finely dispersed fraction  $s_f(r)$ . Figure 3 shows the distributions of the finely dispersed fraction of particles  $s_f(r)$  before (curve 2) and after (curve 1) correction. These distributions were reconstructed from the initial data, presented in Fig. 2. The spectral behavior  $\sigma(\lambda)$  corresponding to the corrected microstructure is shown in Fig. 2 (curve 3). In the example studied the discrepancy at  $\lambda = 0.55 \ \mu m$ dropped from 10 to 0.8% as a result of correction.



FIG. 2. Spectral behavior of the aerosol extinction coefficient  $\sigma(\lambda)$  for  $S_m = 20$  km: 1) model; 2, 3) results of the calculation of  $\sigma(\lambda)$  based on the microstructure s(r), reconstructed without (2) and with (3) correction of the submicron fraction of the haze particles.



FIG. 3. Distribution of the finely dispersed particle fraction of haze before (curve 2) and after (1) correction. The distributions were constructed by inverting the model  $\sigma(\lambda)$  for  $S_m = 20$  km.

## 3. RESULTS OF INVERSION

The final results of the inversion of the spectral dependences of the aerosol extinction coefficient by varying the visibility range  $S_m$  from 5 to 50 km are presented in Fig. 4a.



FIG. 4. The results of inversion of the spectral model of extinction  $\sigma(\lambda)$  of coastal haze: a) size distribution functions of the geometric cross section s(r), reconstructed based on the model  $\sigma(\lambda)$  with the visibility ranges  $S_m = 5(1), 10(2), 20(3), 30(4), and 50$  km (5); b) the total geometric cross section of the particles S(1) as a function of the aerosol extinction  $\sigma(0.55)$  with separation into submicron (2) all coarsely dispersed (3) fractions; c) the volume fill factor V(1) with separation into the submicron (2) and coarsely dispersed (3) fractions as a function of the aerosol extinction coefficient  $\sigma(0.55)$ .

One can see from Fig. 4a that there is a distinct minimum near the point  $r \simeq 0.9-1.0 \ \mu m$  in all reconstructed distributions. This minimum makes it possible to separate the entire range of particle sizes into two regions, corresponding to the finely dispersed  $(r \le 1 \ \mu m)$  and coarsely dispersed  $(r > 1 \ \mu m)$  particle fractions. The curves 1-5 in Fig. 4a describe the microstructure of the haze for values of the meteorological visibility range  $S_m = 5$ , 10, 20, 30, and 50 km, respectively. Figure 4b shows the change in the integral parameters of the microstructure: the geometric cross section of the particles S of the finely and coarsely dispersed fractions (curves 2 and 3) as well as their total magnitude (Fig. 1), as a function of the aerosol extinction coefficient at the wavelength  $\lambda = 0.55 \ \mu m$ . The dependences shown in Fig. 4b are approximated well by linearly increasing functions. When the atmospheric turbidity is increased by a factor of 10 the total cross section increases from 0.03 km<sup>-1</sup> for  $S_m = 50$  km up to 0.3 km<sup>-1</sup> for  $S_m = 5$  km, i.e., also by a factor of 10. In addition, the finely dispersed fraction makes the main contribution to the cross section. Its relative fraction also increases from 63 for  $S_m = 50$  km to 78% for  $S_m = 5$  km as the atmospheric turbidity increases. If we now study the volume content of haze particles, which is characterized by the specific fill factor V, then the relative role of the finely and coarsely dispersed fractions changes (see curves 2 and 3 in Fig. 4c). In this case, conversely, the coarsely dispersed fraction, which constitutes about 78% of the total volume at maximum atmospheric transmission  $(S_m = 50 \text{ km})$ , makes the main contribution. As the atmospheric turbidity increases the relative content of particles of the coarsely dispersed fraction drops over the volume to 69% for  $S_m = 5$  km. The total fill factor (curve 1 in Fig. 4c) is equal to  $3.2 \cdot 10^{-10}$  for maximum atmospheric turbidity ( $S_m = 5$  km) and drops monotonically by approximately a factor of 7 as the visibility range  $S_m$  increases to 50 km. We also note that for the finely dispersed haze fraction the average particle radius over the distribution s(r) ( $r_s = (3/4)$ . (V/S)) decreases from 0.42 to 0.31  $\mu$ m.

Finally, the last microstructural characteristic of interest is the number density of particles. For the haze under study, the particles of the finely dispersed fraction constitute the main fraction of the total concentration of the particles N (more than 99%). The number of particles per cm<sup>3</sup> with radius larger than  $0.09 \ \mu m$  increases monotonically from ~ 70  $(S_m = 50 \text{ km})$  to 1300  $(S_m = 5 \text{ km})$ . Thus depending on the chosen microstructural characteristic of the haze (S, V, or N) the relative significance of its two fractions - submicron and coarsely dispersed changes. It is of interest to estimate the contribution of each fraction to the extinction of optical radiation. Figure 5 shows the results of the calculation of the spectral dependence of the aerosol extinction coefficient for the finely dispersed (curve 1) and coarsely dispersed (curve 2) fractions of marine coastal hazes for the meteorological visibility range  $S_m = 5$  km. The

pattern does not change qualitatively as  $S_m$  increases. As expected, the coarsely dispersed fraction of the haze determines the component with practically neutral spectral behavior in the visible region of the spectrum, where its relative fraction constitutes less than 20%, in the total extinction. The contribution of the coarsely dispersed fraction to the extinction of radiation increases as the wavelength  $\lambda$  increases and becomes dominant for  $\lambda \geq 2.2 \ \mu m$  (at least 90%). It is interesting that in the case of high atmospheric transmission the relative role of the coarsely dispersed aerosol in extinction of radiation increases and its contribution to the total extinction coefficient is not less than 31% (for  $S_m = 50 \ \text{km}$ ).



FIG. 5. The spectral extinction of radiation for the submicron (1) and coarsely dispersed (2) haze particle fractions for the one-parameter model  $\sigma(\lambda)$  (3) for  $S_m = 5$  km.

# 4. CONCLUSIONS

The results of the inversion of the single-parameter model of the extinction coefficient of marine coastal haze, presented in this paper, for the spectral region 0.55-3.9 µm describe the microstructure of aerosol distributions with particle sizes up to 4.3 µm. The inverse-problem method for reconstructing the microstructural distributions of the haze established the existence of the two fractions: a submicron fraction ( $r \le 0.8-1.0 \mu m$ ) and a coarsely dispersed fraction ( $r \ge 1.0 \mu m$ ). This agrees with the modern conception that the structure of atmospheric aerosols is polymodal. The average contribution of the finely dispersed fraction of the haze to the total geometric cross section of the particles is equal to about 70% with a deviation of about 7% as the visibility range varies from 5 to 50 km. The contribution of each fraction to the spectral extinction in the interval 0.55-3.9 µm was estimated. The quantitative information obtained about the dispersion composition of coastal marine haze makes it possible to calculate the different light-scattering characteristics of haze in the visible and IR region, such as, for example, the lidar ratio and the scattering matrix, which are of interest for laser-sounding problems and in other areas of atmospheric optics.

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