Optical-statistical aerosol model of the atmosphere for the Lake Ladoga region

A.V. Vasil'ev and L.S. Ivlev

Research and Development Institute of Physics, St.-Petersburg State University, St. Petersburg

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A microphysical aerosol model has been constructed for the Lake Ladoga region. The model is based on data available from the literature and on the results of airborne measurements of the number density and chemical composition of aerosol particles.

Introduction

Aerosol model of the atmosphere plays a significant part in the problems of numerical simulation of the radiation regime of Lake Ladoga as well as in the inverse problems on determining the atmospheric optical and hydrooptical parameters of the lake.[1,3] Monitoring of its state is important for the city of Saint-Petersburg and the entire Northwest region of Russia.[1,2] Not only mean values but their variances and covariation are also important in solving the above-mentioned problems.

In the case of the aerosol optics, there is a need for a representative bulk of experimental data on the aerosol optical characteristics in order to obtain the aforementioned statistical characteristics. Such measurements have been carried out over Lake Ladoga,[1,4] but it is doubtful to use them for constructing the aerosol optical model, because not the aerosol characteristics themselves were measured but the spectral fluxes and influxes; the measurements were carried out in a narrow wavelength range; statistics of measurements is not sufficiently representative, and so on. So it seems to be the only possible way of solving this problem to apply traditional numerical simulation of the optical properties of atmospheric aerosols. The following models of the microphysical aerosol parameters are assumed: the complex refractive index (CRI), particle size-distribution functions, vertical profiles of the number density, while the optical characteristics are calculated based on these models. In contrast to the measurement data processing, any optical characteristics and in any spectral range can be obtained using this approach.

Since the purpose of a simulation is obtaining of the aerosol optical characteristics, the latter were obtained by varying the aerosol microphysical characteristics. It is easy to construct the models of such variations taking into account strong natural variability of the aerosols. When processing the data of experimental measurements, it is sufficient to determine the rms errors besides the mean values. With the use of literature data and comparing the models of different authors, one can estimate these variations. In constructing the microphysical aerosol model for the region of Lake Ladoga, we used both the literature data and the data of direct airborne measurements of the number density and chemical composition of aerosol particles. The airborne studies were carried out at the Laboratory of Aerosol Physics of the Department of Atmospheric Physics at the R&D Institute of Physics.

Principles of simulation

Aerosol was considered as a random system. Taking into account the fact that our knowledge of aerosol is not complete, we selected the simplest model of statistical variations. For discrete values we set the *a priori* probabilities of occurrence; for continuous values we assumed either uniform distribution over some interval or the normal distribution characterized by the mean value and the variance (the mean value and the covariation matrix for vectors).

Each specific realization of the microphysical model was obtained by means of its simulation (statistical sampling) based on the generation of random values distributed uniformly or normally. Let us introduce the following notations: Rd(a, b) is a random scalar value uniformly distributed over the interval [a, b]; Nd(a, s) is a normally distributed random scalar value with the mean value a and the rms error s; ND(a_i, s_{ij}) is a normally distributed random vector value with the mean value a_i (i = 1, ..., N, N is the vector dimension) and the covariation matrix s_{ij} (j = 1, ..., N).

Technically it is convenient to use the normal distribution when simulating the random values.[5] However, in practice it makes sense, for many parameters, first to set relative variations, second to assume them to be quite large (for example, 100%). But assuming that large variations of some parameters which have the limitations by the physical nature (in particular, positive values) does not match the normal distribution due to its limited nature. For example, negative values appear in approximately 16% of events at the 100% variations. Any way of struggling against them, for example, removing false data, distorts the distribution function by making it asymmetrical, hence, different from the normal one, and simultaneously distorts the real mean value and variance of the random value.

Taking into account that assignment of the relative variation is practically equivalent to the assignment of the variation of the order of magnitude, let us consider the normal distribution of large variations of the limited values. In other words, let us simulate the normal distribution of the random value. On this basis we obtain the algorithm for simulating

$$\mathrm{Ld}(a, d) = \exp\left\{\mathrm{Nd}\left[\ln a/\sqrt{1 + d^2}, \sqrt{\ln(1 + d^2)}\right]\right\},$$

where Ld(a, d) is the lognormal distribution that approximates the normal distribution with the mean value a and the rms error d ($d = s/a$, s is the standard deviation of the normal distribution).

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Taking into account the convention of the model, let us accept the analogous approximation for the multidimensional distribution

$$\mathrm{LD}(a_i, d_i, c_{ij}) =$$
$$= \exp\left\{\mathrm{ND}\left[a_i/\sqrt{1+d_i^2}, \sqrt{\ln(1+d_i^2)\ln(1+d_j^2)}\, c_{ij}\right]\right\},$$

where $\mathrm{LD}(a_i, d_i, c_{ij})$ is the lognormal vector distribution that approximates the normal vector distribution with the mean value a_i, the rms error d_i, and the correlation matrix c_{ij}. Optical characteristics were calculated for each random realization of the statistical microphysical model by the algorithms proposed in Refs. 6–8. The traditional block approach was used in the simulation. The atmosphere was divided into two blocks, the troposphere and the stratosphere.

The tropospheric aerosol was represented in any particular model as a sum of independent aerosol components of different origin. Based on literature data[9,10–13] and the results of direct measurements of the aerosol microphysical parameters, let us select the following blocks of the aerosol model of the troposphere. Two blocks can be selected for Lake Ladoga in winter. They are ice particles and urban aerosol. In summer one can consider four blocks: water particles, urban, soil and marine aerosol.

The effect of stratospheric aerosol on the object under consideration is insignificant as compared with the effect of tropospheric aerosol. So we did not construct any model of the stratospheric aerosol and considered only two blocks,[14] sulfuric acid particles and dust (without distinguishing between volcanic and meteor origin). The aerosol optical characteristics were calculated as a sum of all blocks. For a convenience of calculations,[16] let us approximate the particle size distribution by the single-mode distributions. Then each block was divided into types, i.e., the particles of the same origin but different modes of the distribution. Finally, we obtained the sum of all aerosol types. Each type was characterized by the vertical profile of the number density, CRI of the aerosol substance, parameters of the single-mode size distribution, and (for two-layer particles) the parameters of the inner structure.[18] All the aforementioned characteristics were simulated as random values.

Let us select the following aerosol types in the frameworks of the model. Ice particles over Lake Ladoga in winter and water droplets in summer can be described by a single-mode distribution,[13] the parameters of which were taken the same for ice and water.

Let us use the model proposed in Ref. 14 for the stratospheric aerosol, where the particle size distribution has three modes, the smallest particles are sulfuric acid (sulfuric acid droplets), while the medium-size and coarse particles are dust. Thus, we have three aerosol types in the stratosphere: sulfuric acid, stratospheric dust – 1 and stratospheric dust – 2.

Marine aerosol, i.e., the salt particles produced at evaporation of the marine water sprays, has a two-mode distribution.[11] Then we obtain two types, i.e., the marine salt – 1 and marine salt – 2.

Soil aerosol has a three-mode distribution.[10]

The urban aerosol structure is the most complicated. Urban aerosol is simulated by a six-mode distribution.[12] The first mode is identified as soot, the second one as the organic substance, the third as sulfates, and the last three as dust (let us call them dust – 1, dust – 2, and dust – 3. Taking into account the conventions of the model, let us unify, for simplicity, the urban dust fraction and soil aerosol (soil dust), while taking, at the same time, into account different contribution of these sources in winter and summer.

Thus, we have selected twelve aerosol types. Let us simulate all particles in the spherical shape approximation. Let us take into account aerosol moistening (ice-covering) according to Ref. 8. To take into account moistening of particles in the troposphere and the variations of the sulfuric acid in the stratosphere, we used the statistical models of the vertical profiles of pressure, temperature, and concentration of water vapor.[15]

Models of the CRI of aerosol substances

We obtained the variations of CRI of aerosol substances from the synthesized CRI.[18,18,20] The synthesized CRI was calculated as the weighted mean value

$$m(\lambda) = \sum_{i=1}^{N} p_i\, m_i(\lambda) \Big/ \sum_{i=1}^{N} p_i, \qquad (1)$$

where $m(\lambda)$ is the synthesized CRI (λ is the wavelength); $m_i(\lambda)$ are the CRI of separate components; p_i is the weight of the component; and N is the number of components. The percentage (of mass) of the substance in the mixture composition was used as the weight. Changing the weight p_i, we simulated the variations of the synthesized CRI. Let us use the noted simulation not only for the composite but also for the "simple" substances, by means of synthesizing them from the data of different authors, the data of different authors in Eq. (1) were taken as having the same validity, in other cases the explanations are presented. The CRI were simulated to be the same at different altitudes (except for H_2SO_4 in the stratosphere).

The models of CRI of the aerosol substances used are shown in Table 1. The substances are arranged so that those placed in the upper part of the table can be used as the components of more complex particles. The numbers of remarks presented below are given in the right-hand column of the table.

Table 1. Models of CRI of different substances

Substance	N	Components	p_i	N
Water	3	Refs. 16, 17, 18	Rd(0, 1)	1
Ice	5	Refs. 16, 17, 19, 20, 21	Rd(0, 1)	1
SiO_2	2	Refs. 16, 17	Rd(0, 1)	1
Fe_2O_3	2	Refs. 17, 21	Rd(0, 1)	1
Al_2O_3	1	Ref. 21	1	2
$(NH_4)_2SO_4$	2	Refs. 16, 17	Rd(0, 1)	1
Sulfates	4	$(NH_4)_2SO_4$	Rd(0, 0.5)	3
		$CaSO_4$, Ref. 21	Rd(0, 0.25)	
		$MgSO_4$, Ref. 21	Rd(0, 0.25)	
		Ref. 13	Rd(0, 1)	
Soot	7	Refs. 13, 16, 21	Rd(0, 0.17)	1, 4
		Ref. 18, 4 types	Rd(0, 0.12)	
H_2SO_4	1	Ref. 22	1	5
Organic substance	2	Refs. 10, 16	Rd(0, 1)	1, 6
Stratospheric dust	5	Ref. 16, 3 types	Rd(0, 1)	1, 6
		Ref. 17, 2 types	Rd(0, 1)	
Dust (soil)	9	SiO_2	Ld(0.5, 0.1)	1, 7
		Al_2O_3	Ld(0.15, 0.1)	
		Fe_2O_3	Ld(0.05, 0.1)	
		Sulfates	Ld(0.2, 0.1)	

		Organic substance	Ld(0.1, 0.1)	
		Refs. 13, 16, 17, 23	Rd(0, 0.08)	
Marine salt	3	Refs. 13, 16, 17	Rd(0, 1)	1, 6

1. The list of substances with the same p_i is written in one row in order to reduce the Table size; while actually all p_i were simulated independently.

2. The unique data on CRI of these substances are available at the "collection" of the Laboratory of Aerosol Physics of the R&D Institute of Physics. They were used only in synthesis of CRI of complex substances, so separate statistics of them is not needed.

3. According to the traditional idea, ammonium sulfate is usually assumed to be prevalent when synthesizing the CRI of sulfates. The sulfate model from Ref. 13 was used in addition to the composite model. It was assumed that the models are equivalent.

4. Four types of soot are presented in Ref. 18 that describe strong variations of its CRI depending on the origin.

5. When simulating the CRI of sulfuric acid, the data[22] were used on its dependence on the concentration. Variations of the sulfuric acid CRI were simulated in the frameworks noted in Ref. 22 by varying its concentration. That was determined using the values of temperature and partial pressure of water vapor at certain altitude in the stratosphere.[24]

6. The CRI values synthesized based on the literature data were used for these substances.

7. The model of the dust (soil) composition constructed in Ref. 16 was used in our study. Besides, we used the synthesized models, to which the *a priori* weight of 30% was ascribed in order to retain the soil CRI variation related to the variations of its composition.

Models of the particle size distribution functions

The model from Ref. 23 was used for the stratospheric aerosol, where the generalized lognormal distribution with $b = -3$ was applied to all three types

$$f(r) = \frac{1}{r_0^{b+1}\sigma\sqrt{2\pi}\exp\left[\sigma^2(b+1)^2/2\right]} \times$$

$$\times r^b \exp\left(-\frac{\ln^2(r/r_0)}{2\sigma^2}\right). \qquad (2)$$

Let us take into account the processes of moistening (ice covering) of the tropospheric aerosol. In so doing, let us consider all the parameters to be simulated as the initial nuclei at zero humidity.[8]

We used the lognormal distribution (Eq. (2) with $b = -1$) for pure water and ice nuclei[13,16] and the "marine salt – 1" and "marine salt – 2" types.[11]

The inverse gamma-distribution

$$f(r) = \frac{1}{r_0^{-\nu}\Gamma(\nu)} r^{-1-\nu}\exp(-r/r_0),$$

where $\Gamma(\nu)$ is the gamma-function, was used for the urban and soil aerosols.[12]

Variations of the parameters of the distribution functions were estimated using the data from the aforementioned papers.[9–14,16,23]

Table 2. Models of the particle size-distribution functions

Type	$f(r)$	$m(z)$	$d(z)$, %
Sulfuric acid	r_0 μm	Ref. 23, p. 201	Ref. 23, p. 50
	σ	«	20
Stratospheric dust – 1	r_0 μm	«	Ref. 23, p. 50
	σ	«	30
Stratospheric dust – 2	r_0 μm	«	100
	σ	«	50
Water (ice) nuclei	r_0 μm	0.67	100
	σ	0.9	10
Marine salt – 1	r_0 μm	0.35	100
	σ	0,6	30
Marine salt –2	r_0 μm	1.9	200
	σ	0.8	30
Soot	r_0 μm	0.21	50
	ν	6	50
Organic substance	r_0 μm	0.21	30
	ν	6	30
Sulfates	r_0 μm	1	20
	ν	5	30
Dust – 1	r_0 μm	1.5	30
	ν	5	30
Dust – 2	r_0 μm	3	20
	ν	5	40
Dust – 3	r_0 μm	15	50
	ν	4	60

The distribution function parameters are presented in Table 2. Two rows refer to each aerosol types: $f(r) - r_0$ and σ to the generalized lognormal, and r_0 and ν to the inverse gamma-distribution; $m(z)$ and $d(z)$ are the mean value and rms error of the parameters, respectively. In the general case they are functions of the altitude (see below).

Models of the vertical profiles of the parameters

Vertical profiles of all the parameters were simulated independently. The exponential model[25]

$$\mathrm{corr}(z_i, z_j) = \exp(-|z_i - z_j|/R), \qquad (3)$$

was used for the correlation matrix of the parameters not available in literature. The only parameter to be determined is the correlation radius R.

Let us use the data from Ref. 23 and the model (3) for simulating the vertical profiles of the parameters of the aerosol particle size-distribution functions. Let us select the correlation radius of 3 km on the basis of the characteristic size of aerosol layers in the stratosphere. The height of the tropopause Nd(11 km, 1 km) was taken as the boundary between the stratospheric and tropospheric aerosol. Let us assume that there are no tropospheric aerosol above this boundary and no stratospheric aerosol below it.

As the first approximation, let us ignore the change of the particle size distribution in the troposphere as compared with the vertical behavior of the particle size caused by the humidity height variation. So the parameters of the size distribution of the initial nuclei of the tropospheric aerosol at zero humidity are the same at all altitudes.

The model of the vertical profiles of the parameters of the stratospheric aerosol[23] is presented in Table 3, where $m(z)$ in the mean profile of the number density, $d(z)$ is the

relative rms error of the number density, R is the correlation radius according to Eq. (3), in km.

Table 3. Model of the vertical profiles of the aerosol number density in the stratosphere

Type	$m(z)$	$d(z)$,%	R, km
Sulfuric acid aerosol	Ref. 23, p. 201	200	3
Stratospheric dust – 1	«	200	«
Stratospheric dust – 2	«	300	«

Vertical profiles of water and ice nuclei $C(z)$ were approximated[16] by the formula

$$\begin{aligned} C(z) &= C(0)\exp(-z/d_1), && \text{for } z < z_1, \\ C(z) &= C(z_1), && \text{for } z_1 < z < z_2, \\ C(z) &= C(z_1)\exp(-(z-z_2)/d_2), && \text{for } z > z_2. \end{aligned} \quad (4)$$

The parameters of the model (4) and their variations obtained according to Ref. 16 are presented in Table 4, where m and d are the mean value and the rms error of the parameters, respectively.

Table 4. Model of the vertical profiles of the number density of the water (ice) nuclei

Parameter	m	d,%
$C(0)$, cm^{-3}	20	100
z_1, km	0.5	50
d_1, km^{-1}	0.25	50
z_2, km	3	50
d_2, km^{-1}	1.2	50

Vertical profiles of the aerosol number density over Lake Ladoga and its variations were estimated from the results of direct measurements in this region carried out at the Laboratory of Aerosol Physics. The percentage of aerosols of different origin (marine, urban, soil) was estimated from the experimental data on the aerosol chemical composition. Simulation of the percentage of different types was performed according to Ref. 11 (marine – 1 and marine – 2) and Ref. 12 (six types of the united model of urban and soil aerosols). The correlation radius of all aerosols over Lake Ladoga was assumed to be about 2 km on the basis of the characteristic size of the observed hazes. The mean values of the number density are presented in Tables 5 and 6. The rms errors were estimated according to Refs. 11 and 12 as 100% for "dust – 2" and "dust – 3", and 300% for all other types.

The model of two-layer spheres[8] was used that assumes the growth of the cover depending on the relative air humidity for taking into account moistening (ice covering). It was assumed that the cover consists of water at the positive temperatures and of ice at the negative ones. The empirical parameters determining the growth of the cover and its solubility were also simulated as random values, equal at all altitudes but depending on the aerosol type.

The model was tested while calculating 100 random profiles of the aerosol scattering and absorption coefficients at visible wavelengths. Figure 1 shows the mean vertical profiles at the wavelengths of 0.55 μm. The difference between winter and summer profiles is significant only in the troposphere. The broken character of the profiles over Ladoga in winter is the consequence of the presence of random error in the statistical simulation and can be removed if using larger number of the spectra simulated. However, it is not that essential because the rms error of the profiles presented is 200–300%. Such a rms error is in agreement with the experimental data and theoretical ideas on the aerosol variability in the atmosphere (however, it is directly determined by the simulated rms errors of the number density). Let us note that the range of variation we used includes all conditions of "aerosol weather" from very transparent atmosphere to a dense haze. If the *a priori* data, for example, on the near-ground meteorological visual range, are available, one can select the results of simulation and obtain the mean values and the rms errors corresponding to the specific conditions of observations.

The fact that the volume scattering and especially absorption coefficients over Ladoga have larger values in winter than in summer is explained by a significantly greater portion of soot particles experimentally observed in winter (see Tables 5 and 6). The reason is the slower wash out of soot particles from the atmosphere by precipitation and appearance of additional sources. Let us note that the strong correlation is seen in the figure between the profiles of the scattering and absorption coefficients.

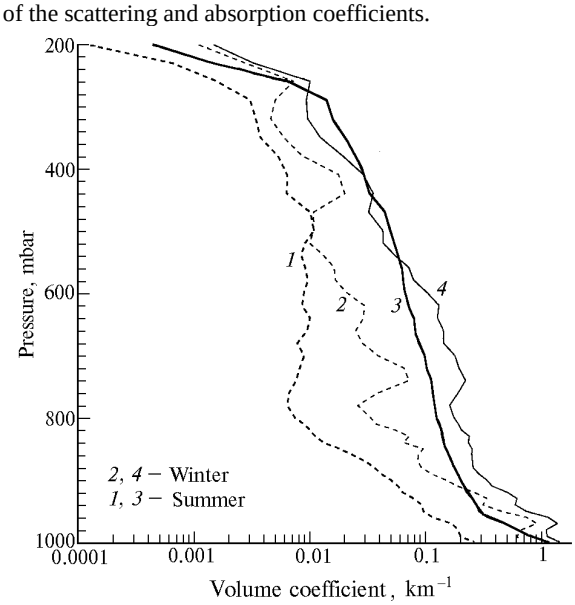

Fig. 1. The mean model profiles of the volume aerosol scattering (*3*, *4*) and absorption (*1*, *2*) coefficients in the troposphere at the wavelength of 0.55 μm.

Table 5. The mean vertical profiles of the number density (cm^{-3}) of the tropospheric aerosols over Lake Ladoga in summer

Height, m	Marine–1	Marine–2	Soot	Organic substance	Sulfates	Dust–1	Dust–2	Dust–3
0	0	0	1500	600	10	10	2	0.02
500	0.6	0.006	500	200	2	5	1	0.01
1000	0.3	0.003	200	60	1	2	0.5	0.005
1500	0.06	0.0006	50	20	0.4	1	0.2	0.002
2000	0.02	0.0002	30	15	0.2	0.5	0.1	0.001
2500	0.02	0.0002	40	20	0.2	0.5	0.1	0.001
3000	0.02	0.0002	40	20	0.2	0.5	0.1	0.001
3500	0.01	0.0001	20	15	0.1	0.4	0.08	0.001
4000	0.01	0.0001	10	6	0.1	0.4	0.07	0.001
5000	0.01	0.0001	10	4	0.05	0.3	0.06	0.0007

6000	0.01	0.0001	10	4	0.05	0.3	0.06	0.0007
8000	0.01	0.0001	10	1	0.01	0.3	0.05	0.0006
10000	0	0	0	0	0	0.3	0.05	0.0005
12000	0	0	0	0	0	0.1	0.02	0.0002
15000	0	0	0	0	0	0	0	0

Table 6. The mean vertical profiles of the number density (cm^{-3}) of the tropospheric aerosols over Lake Ladoga in winter

Height, m	Soot	Organic substance	Sulfates	Dust – 1	Dust – 2	Dust – 3
0	10000	2500	20	6	2	0.003
500	3500	750	6	2	0.5	0.001
1000	1200	250	2.5	1.0	0.2	0.0003
1500	450	100	0.8	0.3	0.1	0.0001
2000	300	75	0.6	0.2	0.05	0.0001
2500	300	75	0.6	0.2	0.05	0.0001
3000	300	75	0.5	0.2	0.05	0.0001
3500	250	50	0.4	0.2	0.04	0.0001
4000	200	40	0.3	0.1	0.03	0
5000	200	40	0.3	0.1	0.03	0
6000	200	40	0.3	0.1	0.02	0
8000	150	30	0.3	0.1	0.02	0
10000	120	25	0.2	0.1	0.02	0
12000	100	20	0.2	0.1	0.01	0
15000	0	0	0	0	0	0

Thus, the results obtained from testing of the model show good agreement between the data of experimental measurements and theoretical ideas on the optical characteristics of aerosol over Lake Ladoga and their variability. This allows us to use the model for solving different problems in optical monitoring of the state of Lake Ladoga.

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