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COMPARISON OF MODEL DISTRIBUTIONS OF AEROSOL PARTICLE SIZES

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Introduction

A model distribution is described by a mathematical expression containing free parameters which are used for fitting the empirical data. These parameters may be related in more or less complex way with common physical entities, e.g., average particle size, etc. A multiplier depending on integral particle concentration is considered as one of the parameters of the model distribution.

In this paper the problem of creating model distributions will be considered without an emphasis on the physical theory of phenomena. It is presumed that the model will be used as a tool for the description of empirical data, and its main criteria of quality are the precision of fitting the data, interpretational simplicity, and mathematical convenience.

Mathematical structure makes it possible to distinguish simple and compound model distributions. Distributions which cannot be reduced to a sum of independently analysable component distributions are called simple. Compound distributions are sums of various simple distributions. The number of parameters of a compound distribution is the sum of the numbers of parameters of its component distributions. The following theoretical considerations are general and concern both, simple and compound distributions. However, the examples deal only with simple distributions.

As a rule, a model distribution with a large number of free parameters, guaranteeing a high precision of curve fitting, is not distinguished by other criteria of quality. Practically, atmospheric physicists have been using simple models with one (distribution a/r^3 of Junge) to six (Smerkalov's distribution [1]) parameters. For different practical tasks there are different optimum model distributions depending on the subset of observations, on the requirements of precision, interpretability, and mathematical convenience. The present paper will study the criteria of quality of model distributions and the application of these criteria in the comparison of some well-known model distributions meant for the description of tropospheric aerosols. A new model distribution (KL-distribution) will be described and investigated.

List of symbols

Continuous distributions will be described using the following symbols:

r - effective, e.g. hydrodynamic, particle radius,

$N(r)$ - concentration of particles with radii below r,

$n_0(r) = dN(r)/dr$ - function of distribution or spectrum of zero order,

$n_p(r) = r^p n_0(r)$ - spectrum of the p-th order,

$M_q(r) = r^q n_0(r)dr$ - $(q-p)$-th moment of the spectrum of the p-th order, e.g., q-th moment of the spectrum of zero order,

$\bar{r}_p = (M_p/M_0)^{1/p}$ - average radius of the p-th order,

$\hat{r}_p$ - modal radius of the p-th order, $n_p(\hat{r}_p) = \max(n_p(r))$.

The most popular tool for the representation of spectra is the function $n_1(r)$, as $n_1(r) = dN(r)/d(\ln r)$.

The model distributions considered

The present paper will investigate only those model distributions which are meant for the description of tropospheric aerosol spectra in wide size range. Expressions of the function $n_1(r)$ are presented in Table 1. For the sake of shortness abbreviations will be used to denote distributions.

Table 1

Model distribution for tropospheric aerosol

Abbreviation	Expression	No of parameters
MG	$ar^\alpha \exp(-br^\beta)$	4
S1A	$ar^{-k} \exp(-br^{-m})$	4
S1B	$a_1(\hat{r}_1/r)^k \exp((k/s)(1-(\hat{r}_1/r)^m))$	4
S2	$ar^{-k} \exp(-b/r - cr)$	4
KLO	$a/((r/r_x)^K + (r_x/r)^L)$	4
KL1	$a_1(K+L)/(L(r/r_1)^K + K(r_1/r)^L)$	4
SME	$a(\exp(-k(\lvert r-\hat{r}_1\rvert/r)^w))/(\hat{r}_1^v + \lvert r-\hat{r}_1\rvert^v)$	6

Table 1 does not include the log-normal distribution and Junge's one-wing distribution a/r^k, as they are used for the description of tropospheric aerosol only in a limited size range.

An analysis of a model distribution usually starts with

the study of its asymptotes, whereas, certain theoretical and empirical premises are taken into account. For instance, according to Smoluchowski's theory of coagulation growth of particles in the conditions of uniform nucleation the left asymptote (small sizes) conforms to the power law. On the other hand, Junge's empirical law points to the power dependence for the right asymptote.

Short comments to the distributions presented in Table 1 are as follows.

MG - well-known modified γ-distribution with power asymptote on the left strongly deviates from Junge's law on the right wing.

S1A - thoroughly described and analysed in [2] by V.I. Smirnov.

S1B - obtained from S1A by elementary transformation of the set of parameters :

$$\hat{r}_1 = (bs/k)^{1/s}, \quad a_1 = a\hat{r}_1^{-k} \exp(-k/s). \qquad (1)$$

The shape of curve and fitting precision by S1A is the same as by S1B and both distributions are called S1. The difference between the two variants becomes evident only in the analysis of stability.

S1 is obtained from MG by inversion of signs of the powers. Therefore the powers in the expressions of MG and S1 are to be considered to be non-negative by definition. S1 has a power asymptote on the right wing which makes it possible to consider S1 as a generalization of Junge's law.

Distributions MG and S1 as special cases of one general distribution were defined by K.S. Shifrin [3].

S2 has been proposed by V.I. Smirnov [4] as a component of a compound distribution.

Distributions KL0 and KL1 are related to each other similarly to the relation of S1A and S1B. Their common denotation in KL. The transformation of the set of parameters is as follows :

$$\hat{r}_1 = (L/K)^{1/(K+L)} r_x, \quad a_1 = aL^{L/(K+L)} K^{K/(K+L)}/(K+L). \qquad (2)$$

L.G. Makhotkin [5] proposed the following estimate of the average spectrum of tropospheric aerosol :

$$n_1(r) = \frac{r^{0.5}}{1 + br^{3.5}}. \qquad (3)$$

This is a good approximation to V.A. Smerkalov's estimate which has been presented in a more complicated form [1]. KL - distribution may be regarded as a generalization of distribution (3).

Model distribution SME has been introduced in [1]. The expression of average spectrum of tropospheric aerosol proposed in the presentation of SME found recognition, but there have been no applications of the distribution itself as a tool of curve fitting. The reason is its evident mathematical inconvenience.

Geometric and analytical properties of KL-model

KL-distribution has power asymptotes both on the left and the right wing. All the parameters of the distribution have a simple interpretation at graphic depiction curve $n_1(r)$ in the logarithmic coordinate grid, presented in Fig. 1.

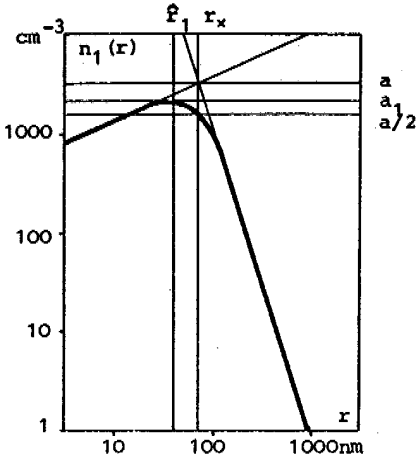

Fig. 1. KL-distribution. a, r_x - coordinates of the intersection point of the asymptotes, a_1, $\hat{r}_1$ - coordinates of the maximum point, L - ascent of the left asymptote, K - descent of the right asymptote. The values of the parameters correspond to Table 2.

Moments of distribution exist when $-L < q < K$ and are expressed in elementary functions :

$$M_q = a\pi r_x^q \;/\; \{(K+L)\,\sin(\pi\frac{L+q}{K+L})\}. \qquad (4)$$

Integral particle concentration:

$$M_0 = a\pi \;/\; \{(K+L)\,\sin(\pi\frac{L}{K+L})\}, \qquad (5)$$

average radii:

$$\bar{r}_p = \{\sin(\pi\frac{L}{K+L}) \;/\; \sin(\pi\frac{L+p}{K+L})\}^{1/p} r_x, \qquad (6)$$

modal radii:

$$\hat{r}_p = \left(\frac{L-1+p}{K+1-p}\right)^{1/(K+L)} r_x. \qquad (7)$$

Distribution KL1 is definite when $K > 0$ and $L > 0$. The scope of KL0 is limited only by the condition $K + L > 0$. Practically this is a considerable advantage of KL0 as one often has to deal with empirical distributions which can be well approximated by KL at negative values of the parameter L.

In atmospheric physics KL-distribution may be used in the size range from some nanometers to the upper limit of the applicability of Junge's law.

Description of the precision of a model

The precision of a model is determined by error of fitting of given spectra using the model. Let us describe a given spectrum with a column vector n which consists of the values of the function $n_p(r)$ over a finite set of radii $r_1, r_2 \ldots$. If the approximation of this spectrum with the model is m , then the error of the approximation is expressed by the vector $e = m - n$. The most natural scalar measure of approximation error is the probabilistic norm

$$\delta^2 = e^T D^{-1} e \;, \qquad (8)$$

where D is the covariation matrix of the errors of measured points of the spectrum curve.

Matrix D is to be given together with the test spectrum n. In the case of description of the precision of the model as

such, it is necessary to determine the conditional standard form of the matrix D. The simplest way is to presume the independence of measurement errors of various points of the spectrum curve and proportionality of errors to the quantities measured. The coefficient of proportionality can have an arbitrary value. Let it be chosen so that $D_{ii} = n_i^2/k$, where k is the number of the values of radii in the representation of the spectrum. Then

$$\delta^2 = \frac{1}{k} \sum_{i=1}^{k} \left(\frac{m_i - n_i}{n_i}\right)^2 \;, \qquad (9)$$

and δ is interpreted as a quadratic mean relative error of approximation.

In the case of the set of J test-spectra, we denote the approximation error of the j-th spectrum through δ_j . If the weights of all test-spectra are the same, then the total estimate of the error will be

$$E^2 = \frac{1}{J} \sum_{j=1}^{J} \delta_j^2 \;. \qquad (10)$$

This quantity is used as an "inverse" measure of the precision of the model.

Loss of measurement information in the interpretation of measurement results

It is said that the aim of measurement is not numbers, but understanding. The part of measurement information used in this understanding could be called useful information while the unused part could be called lost information. The cognitive role of a model in data analysis consists in its power to yield meaningful interpretations of the values of the parameters of the model. In a model representation the measurement information is given by:

- values of the parameters,
- estimates of measurement errors of these values,
- estimates of correlation between measurement errors of the values of different parameters.

A meaningful interpretation uses information given by the values and estimates of measurement errors of the parameters, whereas the information given by the estimates of correlation of errors is, as a rule, lost. This is illustrated by the example in Fig. 2.

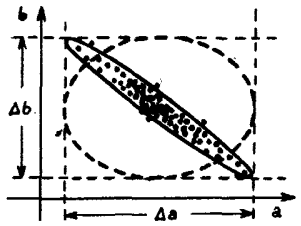

Fig. 2. Results of repeated measurements of the parameters (a, b) of the same spectrum in a bi-parametrical model.

Every dot in Fig. 2 depicts the results of one measurement, the scatter of dots shows measurement errors. Fig. 2 presents simultaneously the ellipse of scattering at a certain confidence level for a two-dimensional measurement result, and the intervals of scattering Δa and Δb separately for every parameter. In the interpretation of measurement results the parameters are considered separately in accordance with their physical content. For instance, parameter a can be the concentration and b - the particle mean radius. The uncertainty of the results is described by the values Δa and Δb ,and the information given by the narrow diagonal form of the ellipse of scattering is lost. This is equal to the replacement of the real ellipse of scattering in Fig. 2 by a wide ellipse shown in Fig. 2 by the indented line. The amount of lost information equals

$$\Delta I = \log \frac{S_2}{S_1} , \qquad (11)$$

where S_2 and S_1 are the areas of the wide and the narrow ellipse. It is easy to demonstrate that

$$\Delta I = \frac{1}{2} \log \frac{1}{1 - R^2} , \qquad (12)$$

where R is the coefficient of correlation between the measurement errors of the parameters a and b .

The same amount of information will be lost, if we increase by K times the measurement errors of every parameter where

$$K = \sqrt{\frac{S_2}{S_1}} . \qquad (13)$$

The quantities ΔI and K are formally equivalent and in practice the one giving a more comprehensible description of the loss of information should be preferred.

Description of the stability of the model

It is said that a model has a poor fitting stability if small disturbances in the approximated spectrum cause large disturbances in the values of the parameters guaranteeing the best approximation. In such cases the ellipse of the scatter of parameter values is strongly elongated, whereas its area may be small. Therefore poor stability is accompanied by large information loss in the interpretation of parameter values. In computational mathematics stability is described by the condition number of the matrix of fitting problem. There are different definitions of the condition number. As there is no expressive interpretation, the use of this concept in our problem is limited. This paper proposes to describe the level of model instability by the amount of information lost in interpretation or by the coefficient of equivalent amplification of measurement errors which are more expressive parameters in our problem.

To generalize the results obtained in the preceding section, let us consider a problem of fitting the spectrum n using the model $m(p)$, where n and m are finite vectors determined on the set of radii r_1 , r_2 , ..., and p is a vector whose elements are the scalar parameters of the model. The dependence of the deviation vector $e = m - n$ on the parameters p could be non-linear. Therefore stability is viewed as a local property depending on the spectrum.

Let us study stability in the neighbourhood of the spectrum $m(p^\circ)$ which is given by certain values of parameters p° . Linear approximation of errors in the neighbourhood of this spectrum is

$$e = B (p - p^\circ) , \qquad (14)$$

where B - is the Jacobi matrix whose elements

$$B_{ji} = \frac{\partial m_j}{\partial p_i} \qquad (15)$$

are calculated in the point p°. Here the index i counts the radii and the index j scalar parameters. The fitting problem is reduced to the task of minimizing the error δ^2 , represented by formula (8) , and it is solved by the well-known methods of the theory of least squares. The solution can be written as:

$$C = (B^T D^{-1} B)^{-1}$$
$$p = p^\circ + CB^T D^{-1} (n - m(p^\circ)) \quad . \qquad (16)$$

Here C - the covariation matrix of parameter values if the spectrum n varies in accordance with the covariation matrix D.

The size of the region of scattering of parameter values calculated by (16), is proportional to $\det C$, which is juxtaposed with the area of the narrow ellipse in Fig. 2. The interval of scattering of the j-th parameter is proportional to C_{jj}. If to ignore the correlations, the size of the scattering region of the parameters will be determined by the product $\prod_j \sqrt{C_{jj}}$ juxtaposed with the area of the wide ellipse in Fig. 2. The respective amount of lost information is

$$\Delta I = \frac{1}{2} \log \frac{\prod_j C_{jj}}{\det C} \qquad (17)$$

and the coefficient of equivalent amplification of errors is

$$K = \left(\frac{\prod_j C_{jj}}{\det C} \right)^{1/(2h)} , \qquad (18)$$

where h - number of scalar parameters.

Other characteristics of the quality of a model

The most important factor of the quality of a model is the cognitive value of its parameters. Unfortunately, there is no way of measuring this factor, and thus expert opinions are decisive.

Practical use of the model depends also on its mathematical convenience. Here three aspects can be mentioned :

1) analytical complexity of the model,
2) complexity and laboriousness of curve fitting using graphic procedures and simple calculations,
3) complexity and laboriousness of a more precise curve fitting usually done by the method of least squares.

There are, again, no exact criteria for the description of mathematical convenience. In this case, however, expert opinions can be supported by some procedures. To estimate analytical complexity, it is necessary to present the expressions of the moments of distributions and characteristic radii. To estimate complexity and laboriousness of an approximation, it is necessary to describe or point out the respective methods and to determine the amount of computation on a standard set of test spectra.

Test spectra for checking the precision and stability of the model

Below in the examples of comparison of model distribution two sets of test spectra are used. The first consists of one spectrum

$$n_1(r) = \frac{a \exp(-0.42\,|1 - 30\mathrm{nm}/r|^{0.47})}{0.000416 + 10^{-6}|r/\mathrm{nm} - 30|^3} \quad , \qquad (19)$$

which is proposed in [1] as the average spectrum of tropospheric aerosols. The second consists of 271 spectra and is obtained as follows. A. Mirme et al. [6] recorded 1661 spectra of tropospheric aerosol near Zvenigorod in the summer 1986. This set has been taken as a basis. All the spectra are presented in a seven-point grid of radii 5, 9, 16, 28, 50, 89, and 158 nm, in the range of this grid the relative measurement error in nearly uniform. 306 spectra turned out to be polymodal and were discarded. The rest were averaged by sets of 5 successively measured spectra. As a result, a set of 207 spectra was obtained, whereas most of them are averages over time interval of about 20 min.

Comparison of the precision of model distributions

The test spectrum (19) has exact presentation in terms of the model SME. The error of fitting of this spectrum with other models is described by Table 2.

Table 2

Model	Approximation of spectrum (19) at $a = 1$	Average relative error
S1A	$2.67\ 10^{15} r^{-4.54} \exp(-36.1 r^{-0.315})$	36%
S1B	$2660 (18.4/r)^{4.54} \exp(-36.1 r^{-0.315})$	36%
S2	$3.06\ 10^{7}\ r^{-2.4} \exp(-38.1/r - 0.00049r)$	70%
KL0	$3100/((r/72)^{3.15} + (72/r)^{0.44})$	7%
KL1	$2140\ 3.59/(0.44(r/42)^{3.15} + 3.15(42/r)^{0.44}$	7%

Table 2 does not contain the model MG which in approximating spectrum (19) is inferior to the model S1. The formal procedure minimizing the error of MG-approximation approaches

the point of inversion where the distribution is not determined and will be transformed into S1.

The results of the comparison of models in the fitting of 271 empirical test spectra are presented in Table 3.

Table 3

Precision of model distributions in the fitting of 271 empirical spectra on a 7-point logarithmic grid of radii from 5 to 158 nm

Model	Relative error average	Relative error maximum	Frequency of turning out as best of compared models
MG	12%	48%	22%
S1	16%	40%	10%
S2	13%	36%	19%
KL	8%	18%	49%

In the execution of computations for Table 3 with MG the limitations $\beta > 0.1$, and with S1 $s > 0.1$, were set up. Without limiting the polarity, the procedure of searching the best fit would "step over" the inversion, and reach S1 in 1/3 of cases, and MG in 2/3 of cases. The limitation of the absolute value was used to avoid losses of computational precision due to the instability of absolute values of the parameter below 0.1.

Comparison of the stability of model distributions

Stability has been studied in the neighbourhood of the best approximations of test spectra (19) described in Table 2. In contrast to precision (which does not change in reversible transformations of the set of parameters of the model) stability is significantly dependent on the representation of the set. Stability is also dependent on the covariation matrix of the disturbances of the spectrum D. Computations have been executed on the assumption of a diagonal structure of the matrix. Let us consider two variants :

- variant of a constant absolute error D_{jj} = const,
- variant of a constant relative error D_{jj} = const $\cdot n_1^2(r_j)$.

The second variant is closer to real situations. Computation results are brought in Table 4.

Table 4

Stability characteristics of the model distributions in the neighbourhood of the average spectrum of tropospheric aerosols according to V.A. Smerkalov, computed on a 13-point logarithmic grid of radii from 5 to 5000 nm.

Model	Constant absolute error ΔI:bit	ΔI:digits	K	Constant relative error ΔI:bit	ΔI:digits	K
S1	11.7	3.5	3.8	8.9	2.7	2.8
S1A	20	6	32	12.6	3.8	8.9
S1B	4.3	1.3	2.1	4.0	1.2	2.0
S1	10.0	3.0	5.6	4.7	1.4	2.2
KL0	3.2	1.0	1.7	3.2	1.0	1.7
KL1	2.0	0.5	1.4	2.2	0.7	1.5

The reasons for poor stability can be studied by analysing the structure of the Jacobi matrix. For instance, in the case of the distribution S1A, the angle between the row vectors, corresponding to the parameters a and s is below 2°. The analysis of the structure of the Jacobi matrix also helps to make proposals for the transformation of the set of parameters in order to build more stable variants of model distributions.

Comparison of computational complexity of model distributions

Methods for the simplified fitting of spectra with the distributions MG and S1 are described in [3] and [2], respectively.

The simplified fitting using the model KL0 can be carried out graphically on a logarithmic coordinate grid (see Fig.1). Straight linear asymptotes are easy to estimate with a transparent ruler. On the condition of uniform relative error the precision of this method is close to that of the numerical method of least squares. An additional guideline to prove the asymptotes is the condition $n_1(r_x) = a/2$. The parameters could be measured as is shown in Fig. 1.

For an exact approximation by the method of least squares the models MG, S1, and S2 are considered on a logarithmic scale, whereas S2 will be linear in reference to all para-

meters, and MG and S1 retain non-linear dependencies on one parameter. The model KL gets the form :

$$1/n_1(r) = pr^K + qr^{-L}, \qquad (20)$$

which retains a non-linear dependence on two parameters. A necessary additional transformation is:

$$r_x = (q/p)^{1/(K+L)}, \quad a = 1/pr_x^K. \qquad (21)$$

The spectrum is to be transformed simultaneously with the estimates of measurement errors or the weights of points of the spectrum by the method of least squares.

In the case of the linear parameters of the transformed model, the computational procedure uses the standard linear algorithm of the least squares method. In the case of the rest of the parameters, methods of the extreme problems are to be used; this takes a considerably greater computational effort. The productivity of an approximation of spectra given in 7 points was empirically determined by means of an Iskra-226 personal computer using a Basic interpreter. The results are presented in Table 5.

Table 5

Productivity of approximation on PC Iskra-226

Model	Spectra per min.
MG	4
S1	4
S2	25
KL	2

The computational complexity of SME is significantly higher than that of the other considered models.

Conclusions

For a comparison of different model distributions it is necessary, on the level of expert decision:

- to estimate the cognitive value of every model which is expressed in the interpretation of parameters;
- to estimate the mathematical convenience of every model;
- to determine the set of test spectra for the calculation of quantitative characteristics of the performance of models.

Characteristics of precision and stability of the model and of laboriousness of the approximation of empirical spectra could be determined on the level of quantitative calculations.

Special attention in the comparison of models should be paid to the estimate of stability, as poor stability brings along not only computational difficulties but, more importantly, large losses of measurement information at the stage of interpretation of results.

A comparison of model distributions of tropospheric aerosols shows that KL-distribution suggested in this paper has the best quantitative characteristics among the considered models.

References

1. Смеркалов Б.А. Аппроксимация среднего распределения аэрозольных частиц по размерам // Изв. АН СССР, ФАО.- 1984.- Т.20, No.4. - С.317-320.

2. Смирнов В.И. Об аппроксимации эмпирических распределений по размерам облачных капель и других аэрозольных частиц // Изв. АН СССР, ФАО.- 1973.- Т.9, No.1.- С.54-65.

3. Шифрин К.С. О вычислении радиационных свойств облаков // Тр. ГГО.- 1955.- Вып.46 (108).- С.5-33.

4. Smirnov V.I. On the mechanisms responsible for the formation of typical size spectra and vertical distribution of tropospheric aerosol particles // Eleventh International Conf. on Atmospheric Aerosols, Condensation and Ice Nuclei.- Budapest, 1984. Vol.1.- P.82-86.

5. Махоткин Л.Г. Многолетний ход величин атмосферного электричества по наблюдениям ГГО // Тр. ГГО.- 1990.- Вып.527.- С.3-7.

6. Мирме А.А., Кикас Ю.Э., Тамм Э.И. Динамика спектра атмосферного аэрозоля приземного слоя // Уч. зап. Тарт. ун-та.- 1988.- Вып.842.- С.109-122.

Translated from:

Таммет Х.Ф. Сравнение модельных распределений аэрозольных частиц по размерам // Уч. зап. Тарт. ун-та.- 1988.- Вып.824.- С.82-108.