MODELING OF THE AEROSOL OPTICAL CHARACTERISTICS IN THE ATMOSPHERIC GROUND LAYER WITHIN 0.3–15 μm SPECTRAL RANGE.
I. PRINCIPLES OF CONSTRUCTING MODELS

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Modeling of atmospheric aerosol optical characteristics is one of the most actively developed directions of atmospheric optics. However, results of these investigations turn out to be contradictory and in some cases troublesome. In our opinion, the reason is a limited number and low reliability of the initial data used for modeling as well as a certain gap in various directions of investigations. Possible ways to overcome the above–indicated intricacies and contradictions are considered in the paper.

Atmospheric aerosols affect actively the atmospheric optical properties within the 0.3–15 μm spectral range. At the same time, their role is essentially different in different ranges of this wide spectral range. In addition to the molecular scattering that is quite stable and can be easy taken into account, the aerosol extinction is the principal factor determining the atmospheric optical characteristics in the visible spectral range for $\lambda < 1$ μm, where absorption bands of atmospheric gases are few and weak or are localized within narrow spectral ranges.

The situation dramatically changes when passing to the infrared (IR) spectral range. Strong absorption bands of water vapor and carbon dioxide, whose central parts practically completely absorb the optical radiation already for optical path lengths of several tens of meters under real atmospheric conditions, for $\lambda > 2$ μm divide this spectral range into a number of atmospheric windows. Numerous absorption bands of trace gaseous components of the atmosphere as well as individual absorption lines and groups of H_2O and CO_2 absorption lines lie within the atmospheric windows characterized by high (on the average) spectral transmission of the atmosphere. So–called continuous water vapor absorption is manifested in the entire range with $\lambda > 2$ μm. It is especially essential in the 8–14 μm atmospheric window, which is of special interest from the standpoint of atmospheric optics in connection not only with its significance for the energy budget of the atmosphere, but also with its wide usage for solving a lot of applied problems. The magnitude of the continuous absorption is comparable to that of aerosol extinction or even more.

Observed in experimental optical investigations is the cooperative effect of all factors. The situation is dramatized by the fact that whereas the measurements of angular and polarization scattering characteristics are feasible in the visible spectral range, i.e., all elements of the scattering phase matrix are available from experimental investigations, in the IR spectral range under real atmospheric conditions only the effects connected with the aerosol extinction and, in some cases, with absorption are observable. It also should be noted that the results of these observations have not very good accuracy and in most cases give only an estimate, because it is difficult to separate correctly the aerosol component from really measured quantity.

Models of optical characteristics of atmospheric aerosol have acquired great significance for correct taking into account the aerosol contribution to the optical properties of the atmosphere. In 1970–80s, i.e., during the comparatively short period when a demand arose for such data, more than 20 models of atmospheric aerosol optical characteristics were proposed (a full list of published papers apart, we refer here only to Refs. 1–3 where a comprehensive overview of such investigations is given). Even the number of the proposed models testifies on the one hand the urgency of the problem and on the other hand the fact that the problem is still far from its solution.

When it comes to constructing an atmospheric aerosol model for optical applications, it is generally implied that such a model must give a set of aerosol optical characteristics corresponding to a given atmospheric state. A specific set of optical characteristics is determined by purely practical needs. In the visible spectral range, it is generally suggested that optical model should provide knowledge of both energy (scattering, extinction, and absorption coefficients), angular, and polarization characteristics. In the IR spectral range, modeling of the only energy optical characteristics is merely required.

Difficulties emerge when determining the set of the parameters that describe the state of the atmosphere and are the input parameters of a model of atmospheric aerosol properties. The specialists in atmospheric optics, realizing the variety and complexity of the processes affecting the optical properties of the atmosphere, have not yet formulated a unified list of conditions that must be monitored in optical observations, if only for comparison of their results.

A more delicate situation exists in atmospheric aerosol researches, because the variability of aerosol properties under real atmospheric conditions (their number density, size distribution, composition of particles, spatial distribution, etc.) is so great that some researchers suggest to consider the aerosol as a random variable.[4] So a common practice of using the meteorological visibility range S_m [or the related aerosol extinction coefficient α ($\lambda = 0.5$ μm) $= 3.902/S_m$] and the relative air humidity $f(\%)$ for input parameters of optical model may be considered as the homage to tradition taking into account practical feasibilities, because these characteristics can be experimentally measured by every possible user of results of modeling. Physically more justified requirements for assignment of

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the aerosol extinction coefficients at some wavelengths or the scattering phase function (its small–angle part) cannot be met in practice. However, the knowledge of these characteristics makes it possible to estimate the aerosol dispersed composition at least approximately.

As practice shows, usage of such a limited set of the input parameters of a model gives quite satisfactory results for models of the ground atmospheric layer in the visible spectral range, but leads to essential contradictions when extending a model to the IR spectral range, where a problem arises of so–called "visually indistinguishable situations", i.e., the situations when the aerosol optical characteristics coincide (or are quite close) in the visible spectral range and differ strongly in the IR range.

Philippov and Mirumyants[5] tried to overcome this problem and introduced rather complex multistep classification of possible situations in the atmosphere. In doing so they continued in essence and tried to improve the first classification of the atmospheric aerosol optical properties.[6] However, the classification proposed in Ref. 6 for the visible spectral range is quite simple (three types of "optical weather" are recognized that differ in the spectral behavior of the aerosol extinction coefficient in the 0.3–1 μm spectral range and are characteristic of the corresponding ranges of variation of S_m and f) and on the average is realistic, whereas the classification proposed in Ref. 5 includes 16 types of optical weather and gives no positive results of any value without their clear–cut recognition. We may say with exaggeration that, first, we must measure the aerosol extinction coefficients in the spectral range up to 15 μm; second, we must recognize the type of optical weather on the basis of classification proposed in Ref. 5; and only then we can apply the results of Ref. 5.

A similar situation takes place when a set of models proposed in Ref. 7 is implemented in practice. It seems that such data are very useful for estimating the large-scale impacts of the atmospheric aerosol on radiation regime of the atmosphere, but the practical applicability of a set of models proposed in Ref. 7 for estimating and predicting the variations of aerosol optical properties attendant to local changes of atmospheric state is doubtful.

Available models of atmospheric aerosol optical characteristics may be divided (obviously, tentatively) into two groups. The first includes the models developed on the basis of statistical processing and generalization of the data of optical measurements in real atmosphere (statistical models). The second group includes the models with the aerosol optical characteristics calculated for different atmospheric situations based on one or another concepts of the structure and microphysical properties of atmospheric aerosols (microstructural models). Among the aforementioned models, the models of Refs. 5 and 6 are related to the first group, because the statistically processed data of direct experimental measurements in specific atmospheric situations are recommended there as model optical characteristics. The models of Refs. 2, 3, and 7 can be considered as an example of models related here to the second group.

An undoubted advantage of the first approach to constructing a model is obvious and vivid relation between model characteristics and really observed quantities. But this advantage of statistical models gives rise to some disadvantages that essentially limit their applicability. In practical implementation of the models, the questions arise first of all of the accuracy of experimental estimates of aerosol extinction and of different formats of representation of the initial data arrays and different procedures of data processing.

The relation with the experimental results is less obvious in the case of microstructural calculation models. First of all, a question arises in their practical implementation or analysis of model results of whether the aerosol model used for calculating the optical characteristics adequately describes the real situation. One more question is the degree of inevitable idealization of a model disperse medium in such calculations. It also should be noted that in this approach the starting problem of prediction of the atmospheric aerosol optical characteristics depending on the atmospheric conditions is replaced by another no less challenging problem of forecast of the aerosol state attendant to changes of atmospheric situation.

This approach to an analysis of model results is impeded by the fact that only the final numerical results of calculations are usually presented. As a result, new experimental data on the complex refractive index of the aerosol matter or new hypotheses on the character of interaction between aerosol and water vapor and so on require all complex of calculations to be made practically from the very start. As a rule, attempts of improvement of a model and its refinement are connected with the change of more than one characteristic of initial model of aerosol microstructure, and in many cases it is difficult to estimate the significance of variation of one or another individual parameter. Taking into account the state–of–the–art concept of physico–chemical properties of atmospheric aerosols, when new experimental data and new ideas appear practically continuously, such work on improvement and refinement of optical models of atmospheric aerosol turns out to be ineffective and in some sense infinite.

It seems more rational to specify a model in the form of the data complex that characterizes in detail the initial model of aerosol structure and composition and a package of algorithms and programs used for calculations rather than in the form of a set of values of some specific parameters. Taking this approach, any potential user of model results can calculate the optical characteristics of interest for the specific values of the input parameters. The most difficult point of this approach is the following. Taking traditional approach, problems associated with the transformation of the model aerodisperse structure under varying conditions (for example, choice of the particle number density and particle size distribution function in various synoptic situations), i.e., in essence, problems of adequacy of the initial microstructural model, have been completely solved by the author of the model, but the ways of its solution, as a rule, are not discussed. The use of the approach declared here means that, though in a few of cases, the user may assign some parameters. In this connection, a complete set of rules and recommendations is obligatory integral part of such a model. Taking into account the state–of–the–art knowledge of the nature and properties of atmospheric aerosols, this part of the model seems to be most difficult for implementation, though, as we will show below when describing the aerosol optical model developed at the Laboratory of Aerosol Physics, certain prospects are really seen in this direction.

There are no technical difficulties in calculating the optical characteristics of a disperse medium now. There are many versions of programs for calculating the polydisperse system optical characteristics on the basis of the Mie theory. However, taking into account the possibility (in a certain sense, the necessity) of practically continuous renewing of the initial data bank as well as the improvement and the refinement of a model, it seems to be very useful to use the principle of modularity proposed in Ref. 8 when constructing the model and the algorithm of its implementation. It may be noted that the special software package OPMODA* has been developed and successfully used in the Laboratory of Aerosol Physics. This package is destined for calculation of the atmospheric optical characteristics, radiative transfer in the atmosphere, and radiation budget of the atmosphere. The principle of modularity was used for constructing this package.

When analyzing the state of the art of problems associated with creation of the optical model of atmospheric aerosol, the following circumstances should be noted:

1) There is a certain gap in the aforementioned approaches that are referred to as statistical and microstructural ones, in other words, in semiempirical modeling based on the experimental optical data and numerical modeling.

2) Results of modeling are contradictory and reasons for the difference observed between the model values are not always clear.

3) Comparison with experimental data is used as a criterion of applicability of the calculated estimates. A great bulk of such data on practically full set of optical characteristics is available in case of modeling of the atmospheric aerosol optical characteristics in the visible spectral range. As was mentioned above, a set of optical characteristics measurable in the IR range is essentially limited, and the results of experimental investigation are difficult for comparison, first, due to the lack of universally accepted concepts of the nature and properties of the H_2O continuous absorption and, second, due to a wide variety of techniques used for its consideration when separating the extinction components.

Using the results of integrated investigations into atmospheric hazes[8–10] carried out during 1971–1978 at the Scientific–Reseach Institute of Physics at the Leningrad State University and at the Siberian Physical Technical Institute (now the laboratory headed by Yu.A. Pkhalagov at the Institute of Atmospheric Optics of the SB of the RAS) we tried on the one hand to reveal the regularities of variation of the aerosol microstructural characteristics and on the other hand to compare the experimental estimates of aerosol extinction with the aerosol optical characteristics calculated on the basis of available experimental microphysical data.

Taking into account the aforementioned, we did the following work.

1) Optical characteristics of individual aerosol fractions were calculated based on the data on microstructure and complex refractive index of particulate matter of individual aerosol fractions (result of laboratory investigations of aerosol samples) taking into account the effect of atmospheric humidity on both aerosol particle size spectrum and optical constants (in this series of papers we discuss only the results related to the energy optical characteristics).

2) The relative contributions from the particle number densities of different fractions were estimated, and the particle size distribution function was refined using the normalized values of the aerosol extinction coefficients by means of comparison with the available experimental data. (As the initial approximation, it was supposed that the particle size distribution function at the relative humidity $f \approx 50–60\%$ obeyed the Junge distribution with $\nu = 3.6$.)

3) The so–refined aerosol particle size distribution functions were compared with the available experimental microphysical data (selectively).

4) The dependence of aerosol particle size distribution function on the parameters characterizing the state of the atmosphere (S_m and f) was parameterized based on the refined aerosol particle size distribution function.

5) The spectral dependence of aerosol extinction coefficients in the IR spectral range and the elements of the scattering phase matrix in the range $\lambda < 1$ μm calculated for the model constructed as described above were compared with the data of independent optical investigations.

Specific contents of individual stages of investigations and their results are considered elsewhere. It should be noted only that the model of aerosol optical characteristics proposed here as a result of our study is, in essence, an intermediate, compromise variant between the traditional approach and that proposed above.

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* The software package OPMODA has been developed by A.V. Vasil'ev

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