Optimizing measurements of size spectra of submicron atmospheric aerosols

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In this paper we propose a method to construct discrete optimal measurement schemes for multimode size-distribution functions of submicron aerosols. The method serves as a basis for numerical simulation of saturated observation plans in the case of a bimodal size-distribution. Some regularities in the formation of optimal screen-type sets are determined. Some reasons for possible instabilities in a solution of the inverse problems for parameters of an aerosol size-distribution are discussed.

Introduction

The accuracy of reconstruction of the parameters of particle size-distribution functions strongly depends on the measurement technique chosen. Problems of optimal planning of experiments as applied to a synthetic screen diffusion battery were discussed in Refs. 1 and 2.

The proposed model of optimization of measurements in the case of the unimodal lognormal distribution has been used as a basis for obtaining the optimal observation plans, as well as their analytical approximations. The number of screens in the obtained plans depends mainly on the geometrically mean radius of particles; it is a monotonically increasing function of this parameter.

In this paper we discuss the capabilities of the proposed algorithm in constructing optimal observation plans for the case of multimode size-distribution functions of atmospheric aerosols.

Significantly increasing dimensionality of the optimization problem of planning observations involves difficulties at direct rounding of simulated continuous plans. In this connection, the procedure of planning is divided into two stages. First a continuous optimal plan is constructed,[3,4] then it is rounded to the discrete one by exhaustion of the calculated spectrum of a continuous plan obtained.

Analysis of results of numerical simulation for a bimodal distribution functions has allowed us to reveal some additional qualitative and quantitative regularities in the distribution of screen-type sets of discrete five-point plans.

Planning of measurements

The aerosol slip, $p(y)$, through y screens, which is measured with the use of a diffusion battery, can be described by the following equation[5]:

$$p(y) = \int_{-\infty}^{\infty} K(y, x)\, f(x)\, \mathrm{d}x,\quad c \le y \le d, \tag{1}$$

where $f(x)$ is the particle size-distribution function of atmospheric aerosol; $x = \log r$, r is the particle radius; $K(y, x)$ is the instrumental function of the following form:

$$K(y, x) = \exp\,(-\,y\, t(x)),$$
$$t(x) = c\,(B_1 \cdot 10^{-x} + B_2 \cdot 10^{-2x})^{2/3}, \tag{2}$$

where $B_1 = 8.53{\cdot}10^{-9}$, $B_2 = 1.27{\cdot}10^{-6}$; c is the constant depending on the velocity of a flow and parameters of the screens.

Equation (1) is a linear integral equation of the first kind for the function $f(x)$. Solution of this equation based on measured values of the function $p(y)$ is an ill-posed inverse problem. Application of mathematical methods for planning experiments under most general assumptions about the form of the distribution function $f(x)$ is rather problematic.[3] Therefore, additional *a priori* information about the properties of the function $f(x)$ is needed.

The planning of observations becomes far more efficient, if the function $f(x)$ is given accurate to the set of parameters. In this connection, from here on we restrict ourselves to presentation of the function $f(x)$ in the form of a sum of lognormal distributions

$$f(x, \theta) = \sum_{i=1}^{m} q_i\, f_i(x, r_{50}^i, \sigma_g^i) \tag{3}$$

with the normalization condition

$$\sum_{i=1}^{m} q_i = 1. \tag{4}$$

Here

$$f_i(x) = (2\pi \log^2 \sigma_g^i)^{-1/2} \times$$

$$\times \exp\{-(x - \log r_{50}^i)^2 / 2 \log^2 \sigma_g^i\},\ i = \overline{1, m}, \quad (5)$$

where r_{50}^i and σ_g^i are the geometrically mean radius of particles and the standard geometrical deviation for the ith mode of atmospheric aerosol; $\theta = (r_{50}^1, \sigma_g^1, \ldots, r_{50}^m, \sigma_g^m)$ is the vector of unknown parameters.

Estimates of the vector θ can be obtained from values of the function $p(y)$ measured at a finite set of points y_k, $k = \overline{1, n}$, $n \geq 3m - 1$, with, for example, the method of least squares. Then, to find numerically the sought vector θ, the procedure of gradient descent can be applied[2,3]:

$$\theta_{j+1} = \theta_j + M^{-1}(\theta_j) \cdot \mathbf{Y}(\theta_j), \quad (6)$$

where

$$M(\theta) = \sum_{k=1}^{n} \sigma_k^{-2}\, \varphi(y_k, \theta) \cdot \varphi^{\mathrm{T}}(y_k, \theta); \quad (7)$$

$$\mathbf{Y}(\theta) = \sum_{k=1}^{n} \sigma_k^{-2}\, \varphi(y_k, \theta)\, \lambda_k;$$

$$\varphi(y_k, \theta) = \nabla_\theta\, p; \quad (8)$$

λ_k is the measured value of the slip-through; σ_k^{-2} is the variance of measurements; T denotes transposition.

It follows from Eq. (6) that the accuracy of estimation depends on characteristics of information matrix (7), which, in turn, depends on the chosen plan of measurement points y_k and their weights s_k, $k = \overline{1, n}$. If we restrict ourselves to consideration of the measurement plans, which maximize the determinant of the information matrix M, then the D-optimal plan can be numerically constructed with the stage-by-stage procedure of sequential analysis and planning of measurements (this procedure is described in Refs. 2 and 3).

In the case of the unimodal distribution function, this procedure has allowed us to simulate numerically the locally optimal plans for the permissible values of the sought parameters. The use of the procedure of planning in the case of a multimodal distribution function faces some difficulties. One of them is difficult rounding of thus obtained continuous plans to the saturated discrete ones, the number of points in which is equal to the number of the sought parameters. In this connection, it is recommended to construct the saturated discrete plan on the spectrum of the continuous plan in the following way[4]:

1. A random sample of size $N = k$ from the spectrum of the continuous D-optimal plan is set.

2. Every point of this sample is sequentially replaced with one of the rest $n{-}k$ points of the spectrum. After each replacement, the determinant of the information matrix of thus obtained plan is calculated; the point bringing maximum value to the determinant is finally included in the plan.

3. Step 2 is repeated with the sample obtained at the previous stage until the determinant of the information matrix increases.

4. These steps are performed with different initial samples from the spectrum of the continuous D-optimal plan; the plan, which suits the criterion of D-optimality best of all, is finally selected.

Numerical experiments

Because calculations are too cumbersome, our consideration is restricted to the bimodal distributions. With the allowance for normalization condition (4), the number of sought parameters is equal to five. In this case, similarly to the case with the unimodal distribution, it is more convenient to pass, in Eq. (3), to a different set of sought parameters:

$$\theta_1 = q_1/\sqrt{2\pi},\ \ \theta_2 = 1/\log \sigma_g^{(1)},\ \ \theta_3 = \log r_{50}^{(1)}/\log \sigma_g^{(1)},$$
$$\theta_4 = 1/\log \sigma_g^{(2)},\ \ \theta_5 = \log r_{50}^{(2)}/\log \sigma_g^{(2)}. \quad (9)$$

Then, taking into account Eqs. (8) and (9), the vector of the basis functions $\varphi(y, \theta) = \nabla_\theta p$ takes a less cumbersome form:

$$\varphi_1 = \int_{-\infty}^{\infty} (\theta_2\, e^A - \theta_4\, e^B)\, dx,$$

$$\varphi_2 = \int_{-\infty}^{\infty} \theta_1\, (1 - \theta_2^2\, x^2 + \theta_2\, \theta_3\, x)\, e^A\, dx,$$

$$\varphi_3 = \int_{-\infty}^{\infty} \theta_1\, (\theta_2^2\, x - \theta_2\, \theta_3)\, e^A\, dx, \quad (10)$$

$$\varphi_4 = \int_{-\infty}^{\infty} (1 - \theta_1)\, (1 + \theta_4\, \theta_5\, x - \theta_4^2\, x^2)\, e^B\, dx,$$

$$\varphi_5 = \int_{-\infty}^{\infty} (1 - \theta_1)\, (\theta_4^2\, x - \theta_4\, \theta_5)\, e^B\, dx,$$

where

$$A(x, y, \theta) = -\, yt(x) - \frac{1}{2}\, (x\theta_2 - \theta_3)^2;$$

$$B(x, y, \theta) = -\, yt(x) - \frac{1}{2}\, (x\theta_4 - \theta_5)^2.$$

The properties of the functions $\varphi_1, \ldots, \varphi_5$ can be studied with the use of numerical simulation. Figure 1 shows the plots of basis functions (10) for a set of possible values of the parameters of lognormal distributions. Analysis of Fig. 1 allows some conclusions to be drawn about the nature of instability in the parameters of the distribution function. This possible instability in solution of the inverse problem is caused by the behavior of the curves $\varphi_1, \ldots, \varphi_5$, because of which information matrix (7) is, in turn, ill-posed. One of the ways to weaken the influence of computational instability is to use optimal screen sets when conducting measurements.

Figure 2 shows the examples of calculation of the optimal schemes of measurements using the algorithm of continuous planning of the experiment followed by best distribution of the number of screens according to the procedure 1–4 of discrete exhaustion of screen sets over the spectrum of the obtained continuous plan.

Numerical simulation of optimal schemes of observations for a bimodal distribution functions allows revealing of additional qualitative and quantitative regularities in arrangement of points in plans. Figure 2*b* demonstrates the step transition of points of the locally optimal plan from one state to another, what is connected with mutual arrangement of geometrically mean radii of particles $r_{50}^{(1)}$ and $r_{50}^{(2)}$.

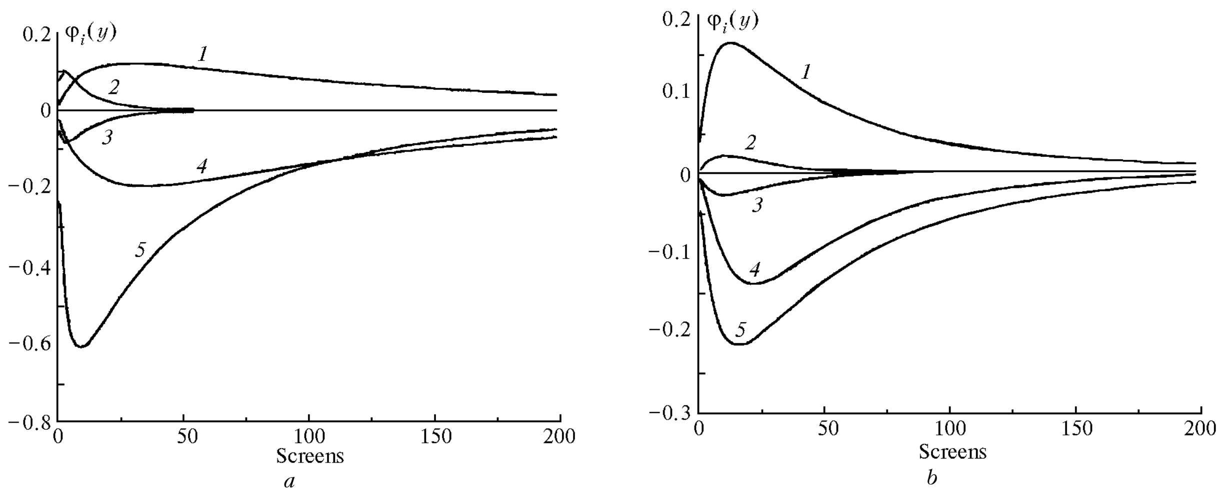

Fig. 1. Basis functions $\varphi_i(y, \theta)$, $i = \overline{1, 5}$, at $q_1 = 0.5$, $r_{50}^{(1)} = 6$ nm, $\sigma_g^{(1)} = 1.7$, $r_{50}^{(2)} = 40$ nm, $\sigma_g^{(2)} = 2.1$ (*a*); $q_1 = 0.27$, $r_{50}^{(1)} = 15$ nm, $\sigma_g^{(1)} = 1.2$, $r_{50}^{(2)} = 20$ nm, $\sigma_g^{(2)} = 1.86$ (*b*).

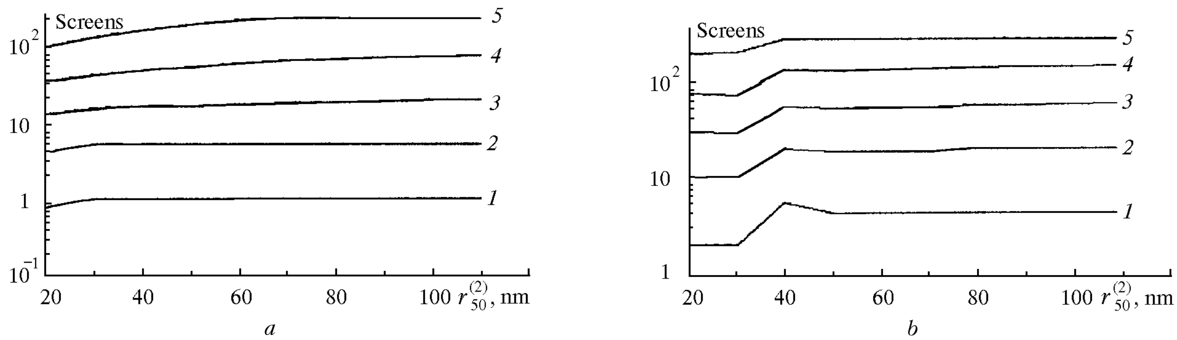

Fig. 2. Discrete locally optimal plans of observations for $\sigma_g^{(1)} = 1.4$, $\sigma_g^{(2)} = 1.86$: $r_{50}^{(1)} = 5$ (*a*) and 15 (*b*) nm.

In the distribution of points of the obtained plans, an exponential growth of the number of screens is observed in the corresponding screen sets. In this connection, it should be noted that the numerically simulated distribution of the number of screens in the screen sets agrees with the recommended distributions, which were obtained empirically.

Conclusion

The conducted numerical simulations of continuous and discrete plans of measurements as applied to the synthetic screen diffusion battery has allowed us to reveal a number of additional peculiarities in their arrangement for the case of the bimodal distribution function as compared to the unimodal one. Numerical analysis of the behavior of the basis functions enables revealing why the inverse problems considered are ill-posed.

The requirements to computational resources have increased significantly. On the one hand, a mutual position of modes in the distribution function may cause a step change in the scheme of observations. On the other hand, the dependence of the position of points in plans on the sought parameters is sufficiently simple. In this connection, it becomes possible to construct analytical approximations of the numerically simulated plans of measurements for widely varying permissible values of the parameters of atmospheric aerosol size-distribution functions.

References

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