OPTIMAL PLANNING IN MEASUREMENTS OF THE SUBMICRON AEROSOL SIZE SPECTRUM WITH A MESH-TYPE DIFFUSION BATTERY

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We propose here a mathematical model of planning the measurements of submicron aerosol size spectrum developed to fit a mesh-type diffusion battery technique in application to the case of an aerosol with a single-mode size-distribution function. The technique proposed enables one to numerically determine an optimal number of meshes. When making computer simulations of locally optimal plans of measurements we have used some realistic parameters of the atmospheric aerosol size spectra. Based on modeling results we have constructed some analytical approximations of the observation plans.

INTRODUCTION

No measurement techniques have so far been developed to cover the entire size spectrum of natural atmospheric aerosols from 10^{-3} and up to 10^{2} μm.[1,2] Therefore, a combination of different methods is usually used with each of these methods being optimal for measurements in a narrower size region.

In recent years a technique of mesh-type diffusion batteries for measuring size of particles in sub-micron region ($d < 1$ μm) is normally used. This technique has first been proposed in the middle 70s (see Refs. 3 to 5). The simplicity of the measuring device design is among its main advantages.

Wide use of this technique in different researches has certainly stimulated a series of theoretical and experimental studies in this area. Those studies were mainly aimed at assessing the limiting capabilities of the approach and estimating the accuracy of the aerosol size-distribution functions that may be obtained in a particular study (see Refs. 3 to 13). In a number of papers the possibilities of the technique to distinguish among multimode distributions have been estimated and limitations of the size-resolution of the measurement technique[14,15] shown.

In Ref.15 it is discussed, based on the results of numerical experiments, that the method only enables a reliable reconstruction of bimodal distributions and it is also shown that there is no any sense in measuring more than six coefficients of the particle breakthrough. However, neither that paper, nor any of the preceding ones did solve the problem on how the concrete parameters of a diffusion battery may influence the measurement accuracy. The problem of choosing the measurement regime regarding the sub-micron aerosol particles obeying a single-mode log-normal size distribution law was considered in detail in Ref. 16 for the channel-type diffusion batteries.

The choice of one or the other way of arranging the measurements can essentially influence the accuracy with which the distribution parameters may be reconstructed. However, the attempts to analyze relevant measurement schemes are very difficult to be performed. This is first of all connected with a multistage research needed and with the difficulty of choosing concrete accuracy criteria when solving the inverse problems of reconstruction and planning the observations. In this paper the problem of planning the experiment based on using a mesh-type diffusion battery (MDB) is considered, and the locally optimal plans of measurements are numerically calculated for the case of a single-mode log-normal size distribution of aerosols.

STATEMENT OF THE INVERSE PROBLEM

The passage of monodisperse aerosol through y meshes of a MDB may satisfactorily be described by the following semiempirical expression[14]:

$$p(y) = \exp(- c\, y\, D^{2/3}(r)), \tag{1}$$

where c is the constant whose value depends on the flow velocity and parameters of the meshes (the cell size and diameter of the threads), r is the radius of particles, in nm, and $D(r)$ is the diffusion factor, which is usually[14] described by the approximate relation which is as follows:

$$D(r) = \frac{B_1}{r} + \frac{B_2}{r^2}, \tag{2}$$

where $B_1 = 8.53\cdot10^{-9}$ and $B_2 = 1.27\cdot10^{-6}$.

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The polydisperse aerosols are normally described by a particle size-distribution function $f(x, \Theta)$. In this case the passage of polydisperse aerosol through y meshes is described as follows:

$$p(y, \Theta) = \int_{-\infty}^{\infty} e^{-y\, t(x)} f(x, \Theta)\, dx, \tag{3}$$

where $x = \log r$; $t(x) = c\, D^{2/3}(r)$, $r = 10^x$, and Θ is the vector of the unknown parameters.

In this paper we consider single-mode size-distributions of particles. The function of particle distribution over size is described, in the majority of aerosol ensembles of condensation and coagulation origin, by the log-normal law,[17] that is, by the expression of the following type:

$$f(x, \Theta) = (2\pi \log^2 \sigma_g)^{-1/2} \times$$
$$\times \exp\{-(x - \log r_{50})^2 / 2 \log^2 \sigma_g\}, \tag{4}$$

where r_{50} is the mean geometrical radius of particles, σ_g is standard geometrical deviation, and $\Theta = (r_{50}, \sigma_g)$.

Let the measured values of the aerosol breakthrough be described as follows:

$$\lambda_k = p(y_k, \Theta) + \xi_k,$$
$$E[\xi_k] = 0, \;\; E[\xi_k \xi_j] = \delta_{kj}\, \sigma_k^2, \;\; k, j = \overline{1, N}\, . \tag{5}$$

Here E denotes the operation of mathematical expectation and δ_{kj} is the Kronecker symbol.

The vector Θ can be estimated using the least-squares method[18]

$$\sum_{k=1}^{n} \sigma_k^{-2} [\lambda_k - p(y_k, \Theta)]^2 \to \min_{\Theta}. \tag{6}$$

Here n is the number of observations, λ_k are the measured values of the aerosol breakthrough through the MDB meshes, and σ_k is the variance of the measurement errors. To improve the accuracy of estimating the vector of parameters, it is worth to optimize, first, the conditions of observations that means that one must properly set a set of the MDB meshes.

To calculate numerically the sought vector Θ, one may apply the procedure of gradient descent[18]

$$\Theta_{m+1} = \Theta_m + M^{-1}(\Theta_m)\, \mathbf{Y}(\Theta_m).$$

Here

$$M(\Theta) = \sum_{k=1}^{n} \sigma_k^{-2}\, \mathbf{f}(y_k, \Theta)\, \mathbf{f}^T(y_k, \Theta)], \tag{7}$$

$$\mathbf{Y}(\Theta_m) = \sum_{k=1}^{n} \sigma_k^{-2}\, \mathbf{f}(y_k, \Theta)\, \lambda_k,$$

$$\mathbf{f}(y, \Theta) = \nabla_\Theta\, p.$$

PLANNING MEASUREMENTS

Let us consider the problem of seeking optimal sets of meshes to provide for identifying the aerosol particle sizes.

Let the plan of observations be understood as the following set of values:

$$\varepsilon_n = \left\{ \begin{array}{cccc} y_1, & y_2, & \ldots, & y_n \\ q_1, & q_2, & \ldots, & q_n \end{array} \right\},$$

where $q_i = S_i / N$, S_i is the number of observations made with the set of meshes y_i, $i = \overline{1, n}$, and $N = \sum_{i=1}^{n} S_i$ is the total number of observations.

In this paper we restrict ourselves, for certainty, to the D-optimal plans, that maximize the information matrix $M(\varepsilon, \Theta)$ determinant, defined by the expression (7).

Strictly speaking, no optimal plan can be constructed *a priori* because of the nonlinear dependence of $p(y, \Theta)$ on Θ**.** Therefore, we have restricted ourselves to the determination of only locally optimal plans. The determination is being done using the following procedure of the sequential analysis and planning of the observations.[18]

1. Let the experiment be represented by $K - 1$ observations performed following a nondegenerate plan ε_{K-1} (i.e., $|M(\varepsilon_{K-1}, \Theta)| \neq 0$). Let us find the point y_K such, that

$$d(y_K, \varepsilon_{K-1}, \Theta_{K-1}) = \max_{y \in \Omega} d(y, \varepsilon_{K-1}, \Theta_{K-1}),$$

where

$$d(y, \varepsilon_{K-1}, \Theta_{K-1}) =$$
$$= \mathbf{f}^T(y, \Theta)\, M^{-1}(\varepsilon_{K-1}, \Theta)\, \mathbf{f}(y, \Theta)\, |_{\Theta = \Theta_{K-1}}.$$

2. Let an additional observation $\lambda_K = p(y_K) + \xi_K$ be done at the point y_K.

3. Then we seek the estimates, according to the least squares method, of Θ_K from K the observations following the plan:

$$\varepsilon_K(y_K) = \frac{K-1}{K}\, \varepsilon_{K-1} + \frac{1}{K}\, \varepsilon(y_K),$$

where $\varepsilon(y_K)$ is the single-point plan.

After the operations performed following the point 3, we return again to the operations under point 1, and so on, while

$$|M^{-1}(\varepsilon_N, \Theta_N)| / N$$

will not become less than some defined value.

Note 1. If there are approximate estimates of r_{50} and σ_g available, the procedures 1–3 can be replaced by the following two ones[19]

– to construct an optimal plan ε and to carry out the observations according to it ;

– to estimate r_{50} and σ_g, using the measurement performed according to the plan ε.

Note 2. In the case of a monodisperse aerosol and the regression (1) the locally optimal single-point plan can be represented in an explicit form as the function of particle radius.

The matter is that in this case the optimal number of meshes is determined by the condition when the below expression reaches its maximum

$$M(\varepsilon, r) \equiv \left(\frac{\partial p}{\partial r}\right)^2 \to \max_{\varepsilon}. \quad (8)$$

In view of the necessary condition of the function extremum (8) over y, it is simple to obtain an unknown quantity of the point of the optimal plan

$$y = 1/[cD^{2/3}(r)]. \quad (9)$$

NUMERICAL EXPERIMENTS

Strictly speaking, no optimal plan can be constructed *a priori* because of the nonlinear dependence of the regression function (3) on the unknown parameters r_{50} and σ_y. Therefore in this section, using the procedure 1–3, we restrict ourselves to the numerical simulation of locally optimal plans of observations for the set values of the vector $\mathbf{\Theta}$.

The expression (3) and it derivatives with respect to the relevant parameters can essentially be simplified if making use of the following substitution:

$$\omega_1 = \frac{1}{\log \sigma_g}, \qquad \omega_2 = \frac{\log r_{50}}{\log \sigma_g}. \quad (10)$$

Then, taking into account Eqs. (3), (4), and (10), we obtain

$$p(y, \omega) = (2\pi)^{-1/2} \int_{-\infty}^{\infty} \omega_1 \, e^{A(x,y,\omega)} \, dx,$$

$$\frac{\partial p(y)}{\partial \omega_1} = (2\pi)^{-1/2} \times$$

$$\times \int_{-\infty}^{\infty} (1 - \omega_1^2 x^2 + x\omega_1 \omega_2) \, e^{A(x,y,\omega)} \, dx,$$

$$\frac{\partial p(y)}{\partial \omega_2} = (2\pi)^{-1/2} \times \int_{-\infty}^{\infty} \omega_1 (x\,\omega_1 - \omega_2) \, e^{A(x,y,\omega)} \, dx,$$

where

$$A(x, y, \omega) = -yt(x) - \frac{1}{2}(x\,\omega_1 - \omega_2)^2.$$

Let us take as the characteristic range r_{50} variation the interval of the aerosol particle sizes from 5 to 200 nm, while the standard geometrical deviation σ_g being from the interval (1.2; 2.5).

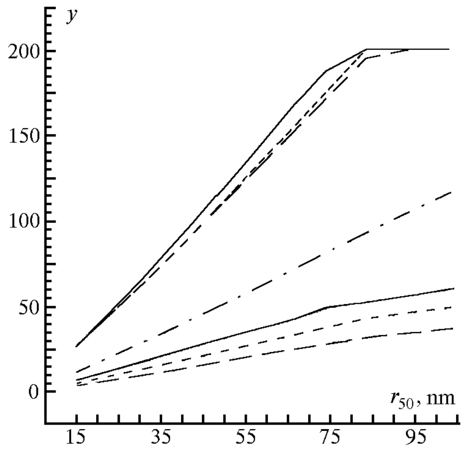

FIG. 1. The lower and the upper number of meshes in the locally optimal plans: $\sigma_g = 1.2$ (solid line), $\sigma_g = 1.5$ (dashed line), and $\sigma_g = 1.8$ (long–dashed line), dot–and–dash line is the position of the optimal planning point as a function of the particle radius in the monodisperse case.

Figure 1 shows the results of simulation of the local D-optimal plans for the fixed values $\sigma_g = 1.2$, 1.5, and 1.8 and the current values of r_{50} from the area of admissible values taken with the step of 5 nm. The plans constructed consist of two sets of the meshes, $y_1(\omega)$ and $y_2(\omega)$, that include different number of meshes. As is seen from the figure, the number of meshes in the plans obtained is mainly determined by the geometric mean radius of particles being monotonically increasing functions of r_{50}.

At $r_{50} \approx 85$ nm the functions $y_2(\omega)$ break and degenerate into a straight line, which is parallel to r_{50}. In this case it is connected with the limitation on the number of meshes admissible for use.

The relations of the optimal plan points to r_{50} and σ_g shown in the figure enable us to propose quite good analytical approximations. The position of the measurement points from the plans is quite satisfactorily described by the following dependences:

$$y_1(\Theta) = \cot(c_1\,\sigma_g)\, r_{50} + a_1,$$

$$y_2(\Theta) = \begin{cases} \cot(c_2\,\sigma_g)\, r_{50} + a_2, & \text{at } r_{50} < 85 \text{ nm}, \\ 200 \text{ grids}, & \text{at } r_{50} \ge 85 \text{ nm}, \end{cases}$$

where $c_1 = 0.3$, $c_2 = 0.7$, $a_1 = -10$, and $a_2 = -2$.

CONCLUSION

In this paper we have analyzed the problem on optimizing the measurement conditions when studying the atmospheric aerosol that is described by a single-mode size-distribution function. The mathematical methods of planning the experiment proposed in this paper, may well be applied to the cases when multimode distribution functions, if taking into account some additional relationships.

The efficiency of planning the observations strongly depends on the vast complex of the conditions to be fulfilled. First of all the mathematical model used must be adequate to the level of its certainty required. The possibilities that the system of observations may provide are also very important. On the other hand, the optimal plans of measurements constructed allow us to formulate in a certain sense the limiting conditions of the inverse problem stability.

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