

## ANALYSIS OF MEASUREMENT ERRORS IN AEROSOL DISPERSE COMPOSITION AND CONCENTRATION FOR TIME-OF-FLIGHT TYPE PARTICLE ANALYZERS

V.A. Yashin, Yu.D. Makashev, and V.S. Toporkov

*Scientific Research Institute of Aerobiology,  
State Scientific Center V" "Vector", Novosibirsk Region*

*Received February 4, 1998*

*The problem of space coincidence of aerosol particles in the sampling volume of laser time-of-flight particle analyzer has been studied. Using model distributions, it is shown that particle coincidence in space may lead to errors in determining aerosol disperse composition and concentration. The retrieval of parameters of the initial aerosol is shown to be ambiguous when the inverse problem is solved. Possibilities of improving measurement accuracy and stability are discussed.*

Photoelectric particle counters (PEPCs)<sup>1</sup> and time-of-flight laser particle analyzers (TFLPAs)<sup>2</sup> have recently become widely used in aerosol studies. Among the advantages of the latter are high resolution, wide size range, and possibility of conducting automated measurements in real-time. However, one of the shortcomings of these devices is that there exists a nonzero probability that two or more particles may simultaneously occur in the sampling volume and coincide in space (the coincidence problem). Depending on particle analyzer design, two such particles can be counted as a single one, if at all, what leads to a sampling error.

Recently, the coincidence problem has been studied rather well for PEPCs,<sup>1,3</sup> but these results can hardly be used for TFLPAs having different measurement setup. The PEPCs measure scattered intensity from particles moving in the sampling volume with equal velocities, so that the probability of their coincidence is determined only by the total particle concentration. On the contrary, TFLPAs measure particle velocity while those, after being accelerated in a nozzle, cross the sampling volume with different velocities determined by their aerodynamic diameter. Thus, the probability that two or more particles coincide in the TFLPA sampling volume depends not only on particle concentrations, but on the particle sizes either while that is ignored in the PEPC theory.

The present paper focuses on the study of errors in determining aerosol concentration and disperse composition due to particle coincidence in the TFLPA sampling volume. The counting chamber has the shape of a cylinder 0.7 mm in diameter and 0.25 mm in height. The electronic circuit is designed to record only those particles that enter the empty sampling volume provided no other particles enter it during the sampling time. In the initial flow of  $n_0(x_i) \Delta x$  particles in the size range from  $x_i$  to  $x_i + \Delta x$ , the device records  $n(x_i) \Delta x$  particles in this size range:

$$n(x_i) \Delta x = n_0(x_i) \Delta x P_0 P_1, \quad (1)$$

where  $P_0$  is the probability of a particle entering empty sampling volume, and  $P_1$  is the probability that no more particles enter the sampling volume after a given particle entered it.

The probability of a particle entering the empty sampling volume depends only on the total concentration<sup>3</sup>:

$$P_0 = \exp(-N_0 V), \quad (2)$$

where  $N_0$  is the total particle concentration of the initial aerosol, and  $V$  is the volume of the counting chamber.

To derive  $P_1$ , we used the results of Ref. 4, where it is found for the PEPC that

$$P(m, \tau) = \exp[-(N_0 V + m) \tau]; \quad (3)$$

$$\tau = u t / h, \quad (4)$$

where  $P(m, \tau)$  is the probability that  $m$  particles are detected in the sampling volume with time, provided that initially there were  $m$  particles in it;  $u$  is the particle velocity;  $h$  is the linear size of the counting chamber in the direction of particle flow; and  $t$  is time.

If we divide the entire size range into equal intervals  $\Delta x$  and assume that all particles in a given interval have the same velocity  $u(x_i)$  at the nozzle exit, then we can calculate the probability  $p_{ij}$  that, given the particle  $x_i$  is initially in the sampling volume, no other particle  $x_j$  will enter the sampling volume over the time  $t(x_i)$  required for particle  $x_i$  to traverse the sampling volume, namely

$$p_{ij} = \exp \left[ - \frac{n_0(x_j) \Delta x V u(x_j) t(x_i)}{h} \right] \quad (5)$$

for  $i \neq j$ ; and

$$p_{ii} = \exp \left\{ - \frac{[n_0(x_i) \Delta x V + 1] u(x_i) t(x_i)}{h} \right\} \quad (6)$$

for  $i = j$ .

Since

$$u(x_i) = h/t(x_i), \quad (7)$$

Eqs. (5) and (6) become

$$p_{ij} = \exp \left[ - \frac{n_0(x_j) \Delta x V u(x_j)}{u(x_i)} \right] \quad (8)$$

for  $i \neq j$ ; and

$$p_{ii} = \exp \left\{ - \frac{[n_0(x_i) \Delta x V + 1] u(x_i)}{u(x_i)} \right\} \quad (9)$$

for  $i = j$ .

Assuming the non-interacting particles of a system to be randomly located in space, we can write

$$P_i = \prod_{j=1}^{\infty} p_{ij}, \quad (10)$$

where  $P_i$  is the probability that, given the particle  $x_i$  is initially in the sampling volume, the particle  $x_i$  will be detected in a time interval  $t(x_i)$ .

Proceeding to the limit  $\Delta x \rightarrow 0$  we have for  $P_1$

$$P_1 = \lim_{\Delta x \rightarrow 0} P_i, \quad (11)$$

$$P_1 = \exp \left[ - \frac{V}{u(x_i)} \int_0^{\infty} u(x_j) n_0(x_j) dx_j - 1 \right]. \quad (12)$$

Using Eqs. (1), (2), and (12) with subscripts  $i$  and  $j$  omitted, we arrive at the dependence of the measured disperse composition and concentration on the initial aerosol parameters of the following form:

$$n(x) = n_0(x) \exp [-(N_0 V + 1)] \times \exp \left[ - \frac{V}{u(x)} \int_0^{\infty} u(x) n_0(x) dx \right]; \quad (13)$$

$$N = \int_0^{\infty} n_0(x) \exp [-(N_0 V + 1)] \times \exp \left[ - \frac{V}{u(x)} \int_0^{\infty} u(x) n_0(x) dx \right] dx. \quad (14)$$

Equations (13) and (14) can be used to predict errors in determining aerosol concentration and disperse composition, provided that the electronic circuit operates as described above. As calculations showed, the concentration retrieval is uncertain because of uncertainty in the shape of the initial aerosol size distribution. Exact shape of the curve of measured concentration versus initial concentration depends on the type and parameters of the initial aerosol size distribution (Fig. 1).

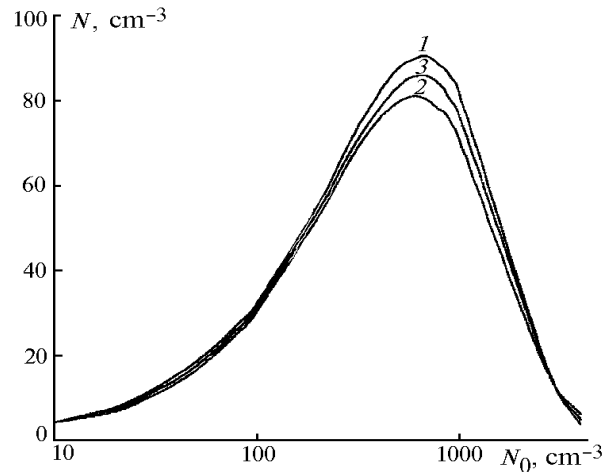


FIG. 1. Measured concentration versus initial concentration for different types of the initial distribution: monodisperse distribution with  $n_0(x) = 1$  for  $x = 10 \mu\text{m}$  and  $n_0(x) = 0$  elsewhere (curve 1); rectangular distribution with  $n_0(x) = 0.1$  for  $1 \mu\text{m} \leq x \leq 10 \mu\text{m}$  and  $n_0(x) = 0$  elsewhere (curve 2); and lognormal distribution with  $D_{50} = 10 \mu\text{m}$  and  $\sigma_g = 2.25$  (curve 3).

It should be noted that this dependence is quite weak. As the initial distribution changes from monodisperse to polydisperse (uniform) distribution, the position of the maximum and its amplitude change only by 10–15%.

Errors in determining disperse composition (Fig. 2) arise from uncertainty in both the initial concentration and parameters of the initial aerosol distribution.

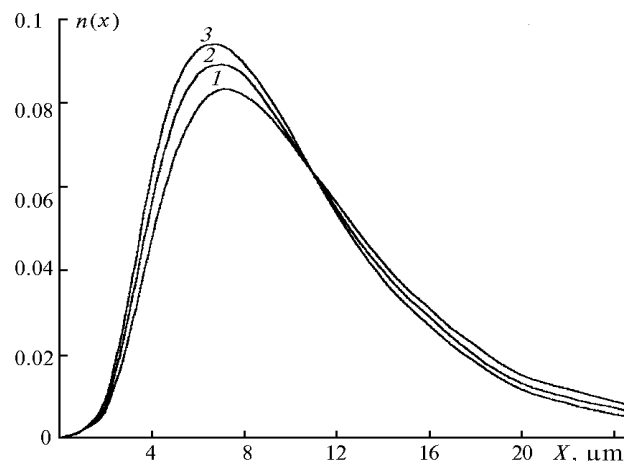


FIG. 2. Error in the disperse composition retrieved assuming the initial lognormal distribution with  $D = 10 \mu\text{m}$  and  $\sigma = 1.75$  at different concentrations: initial distribution (curve 1); calculated distribution for the initial concentration  $N_0 = 10^3 \text{ cm}^{-3}$  (curve 2); and, calculated distribution for the initial concentration  $N_0 = 2.5 \cdot 10^3 \text{ cm}^{-3}$  (curve 3).

Also of great interest is to solve the inverse problem, i.e., to reconstruct the initial aerosol concentration and disperse composition from the measured ones. For this aim, we recast expression (13) into the form

$$n_0(x) = n(x) \exp(N_0 V + 1) \times \exp \left[ \frac{V}{u(x)} \int_0^\infty u(x) n_0(x) dx \right]. \quad (15)$$

Further, multiplication of both its sides by  $u(x)$  and integration over particle size yields

$$z = \int_0^\infty u(x) n(x) \exp(N_0 V + 1) \times \exp \left[ \frac{V}{u(x)} z \right] dx, \quad (16)$$

$$z = \int_0^\infty u(x) n_0(x) dx. \quad (17)$$

Finally, upon integration of both sides of Eq. (15) over particle size, and taking into account Eqs. (16) and (17) we obtain the system of equations for two unknowns,  $z$  and  $N_0$ :

$$z = \exp(N_0 V + 1) \int_0^\infty u(x) n(x) \times \exp \left[ \frac{V}{u(x)} z \right] dx; \quad (18)$$

$$N_0 = \exp(N_0 V + 1) \int_0^\infty n(x) \times \exp \left[ \frac{V}{u(x)} z \right] dx. \quad (19)$$

This system was solved numerically using a computer. We first processed the initial size distribution with the "Prognoz" program and the resulting distribution was then used to solve the inverse problem. As a result, for the system of equations (18) and (19), we have obtained two solutions,  $z_1, N_{01}$  and  $z_2, N_{02}$ , and, subsequently, two different aerosol size distributions.

One solution recovers the initial size distribution, as expected; while the other one gives a new distribution, which differs from the initial one (Fig. 3). It is interesting that the two distributions differ not only by the concentration, as for PEPC measurements, but also by the disperse composition, while, when measured by TFLPA, they produce identical results. For both initial distributions, there is a concentration at which the ambiguity disappears. This is the point of maximum on

the curve of measured concentration versus the initial concentration (see Fig. 1).

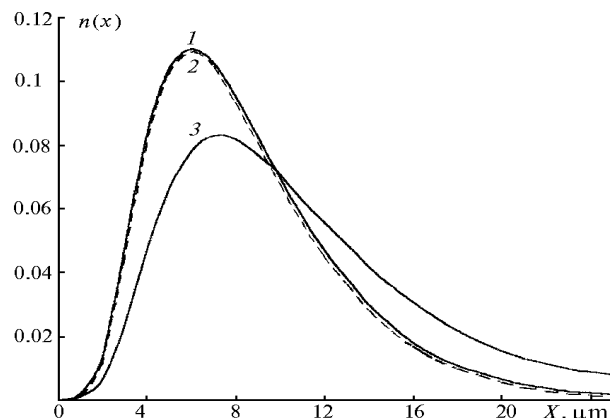


FIG. 3. Solution of the Inverse problem: initial distribution (curve 1), first solution (curve 2), and second solution (curve 3).

Theoretical studies show that particle coincidence in the TFLPA sampling volume leads to errors in the aerosol concentration and disperse composition measured. The errors may be very large, especially when the initial aerosol concentration is high. When the inverse problem is solved, uncertainty arises not only in the retrieved aerosol concentration, but also in the aerosol disperse composition, which represents an additional source of uncertainty in practice. This difficulty can be avoided, in particular, by limiting the aerosol measurements to concentration range, where the errors in aerosol disperse composition are insignificant.

Scientific Research Institute of Aerobiology at State Scientific Center V "Vector" has constructed the instrumentation complex consisting of TFLPA and concentration-limited disintegrator for studying dry powders. The concentration limit is maintained using a signal from TFLPA with a negative feedback at the level of about  $5 \cdot 10^2 \text{ cm}^{-3}$ .

The experiments that have been carried out show the efficiency of the developed instrumentation in increasing the measurement accuracy.

### REFERENCES

1. S.P. Belyaev, N.K. Nikiforov, V.V. Smirnov, and G.I. Shchelchikov, *Opto-Electronic Methods for Aerosol Study* (Energoizdat, Moscow, 1981), 232 pp.
2. T.C. Wilson and B.Y.H. Liu, *J. Aerosol Sci.* **11**, No. 2, 139–150 (1980).
3. K. Willeke and B.Y.H. Liu, in: *Fine Particles*, ed. by B.H.N.Y. Liu (Academic Press, 1976), pp. 697–729.
4. J.V. Julianov and A.A. Lushnikov, *J. Aerosol Sci.* **15**, No. 1, 69–81 (1984).