Corrections for the Effects of Particle Trajectory and Beam Intensity Profile on the Size Spectra of Atmospheric Aerosols Measured with a Forward Scattering Spectrometer Probe

YOUNG J. KIM* AND JOE F. BOATMAN

Air Quality Group, Environmental Research Laboratories/NOAA, Boulder, Colorado

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ABSTRACT

Distortion of the size spectra measured with a forward scattering spectrometer probe (FSSP) under different transit time modes—"inhibit", "normal", and "delayed"—was evaluated using the theoretical analyses by Baumgardner and Spowart and the results of the response time and beam intensity profile measurements of the NOAA FSSP. The Baumgardner and Spowart work is extended to correct the FSSP atmospheric aerosol data collected under the "inhibit" or "delayed" mode. A correction algorithm is developed using the non-negative least squares (NNLS) method to reconstruct the original size distribution from a distorted one measured with an FSSP under the inhibit or delayed mode. A lognormal fit to the corrected size spectra was able to successfully recover from the original size distributions from the distorted artificial ones obtained from the theoretical simulation of the FSSP performance. When the actual test flight data for atmospheric aerosols measured with the NOAA FSSP under the inhibit and delayed modes were corrected using the NNLS correction scheme, the two corrected size spectra converged, implying the measurement of the same sample of particles.

1. Introduction

The Air Quality Group of the Environmental Research Laboratories, National Oceanic and Atmospheric Administration (NOAA/ERL) has been conducting various field measurements for the past several years, which involve airborne measurements of atmospheric trace species (Boatman et al. 1989). During those field measurements, the size distribution of atmospheric aerosols was measured using two Particle Measuring Systems, Inc. (PMS) optical spectrometers, an active scattering aerosol spectrometer probe (ASASP) and a forward scattering spectrometer probe (FSSP), mounted on the wings of the NOAA King Air research aircraft (Kim et al. 1988). The FSSP was initially designed for in situ measurements of cloud droplet size distribution (Knollenberg 1976). Advent of an in situ probe such as an FSSP has greatly contributed to a better understanding of the microphysics of clouds and aerosols. However, measurement problems associated with the FSSP, due to its inherent optical and electronic limitations, are well known and have been investigated to date by many workers (Pinnick et al. 1981; Cerni 1983; Dye and Baumgardner 1984; Baumgardner et al. 1985, 1986; Baumgardner 1987; Cooper 1988; Baumgardner 1988; Baumgardner and Spowart 1990). The response time of the FSSP electronics affects the measurement of droplet size and concentration (Baumgardner et al. 1986). Optical coincidence and electronic dead-time can also cause undercounting of particles (Baumgardner et al. 1985; Cooper 1988).

Undercounting errors due to optical coincidence and electronic dead-time can be neglected for atmospheric aerosol measurement applications where number concentrations of coarse particles are about two orders of magnitude lower than inside a cloud layer. One problem with an FSSP which needs to be investigated is the sizing error due to the effects of particle trajectory through an optical beam cross section having a nonuniform intensity distribution. The FSSP utilizes the Velocity Reject feature by rejecting particles in the depth-of-field (DOF) if their transit time does not exceed the running average of all previous particles' transit time. Two transit time modes of operation are available for an FSSP in determining the transit time of each signal pulse. In "normal" mode the transit time is measured between minimum threshold points corresponding to the first PHA level. In "delayed" mode the transit time is measured between the half maximum points of each voltage pulse. The delayed mode is recommended by the manufacturer for measuring the size distribution at sampling speeds above 25 m s^{-1}. When

* Cooperative Institute for Research in Environmental Sciences, University of Colorado.

Corresponding author address: Dr. Young J. Kim, R/E/ARx1, Aerosol Research Section, NOAA/ERL, 325 Broadway, Boulder, CO 80303-3328.

the "inhibit" mode is selected, the velocity rejection circuitry is turned off. Thus, all particles in the DOF are accepted regardless of their transit time.

The effects of the electronic response time limitations and the laser beam intensity distribution on the FSSP measurements were previously investigated (Baumgardner 1988; Baumgardner and Spowart 1990). A correction method was reported which was able to minimize those effects for the cloud droplet size spectra measured under normal mode (Baumgardner 1988; Baumgardner and Spowart 1990). The main impetus of this paper was to extend the analyses of Baumgardner and Spowart (1990) to develop a correction algorithm which can reconstruct the actual size distributions of atmospheric aerosols from distorted size spectra measured with an FSSP under different transit time modes. A numerical inversion algorithm based on the nonnegative least squares (NNLS) method is developed in this paper for correction of the FSSP data measured under the inhibit or delayed mode. This correction algorithm was successfully applied to hypothetical lognormal distributions as well as actual test flight data, resulting in satisfactory deconvolution of the original distributions.

2. Evaluation of operational characteristics of an FSSP

The FSSP uses a multimode He–Ne laser as a light source in order to have as uniform an intensity distribution as possible across the laser beam cross section. However, the laser beam intensity distribution of the NOAA FSSP probe was measured to be nonuniform, having a maximum intensity at its center region (see Fig. 1). The intensity of the laser beam was measured using the method of Baumgardner and Spowart (1990)

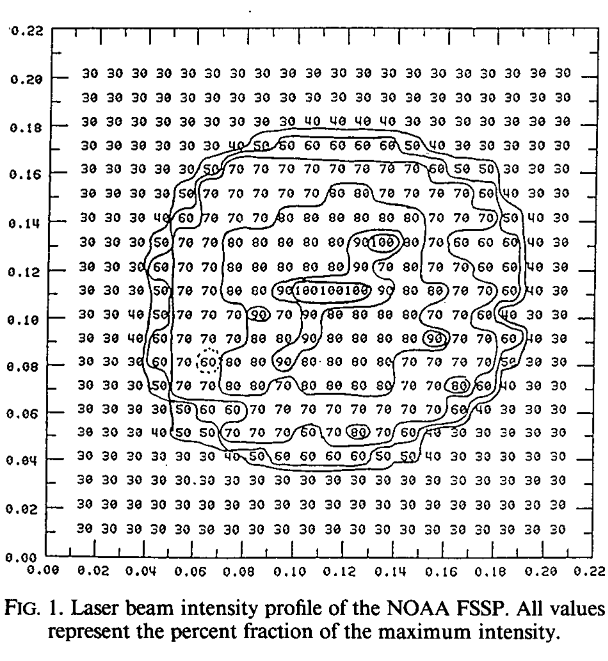

FIG. 1. Laser beam intensity profile of the NOAA FSSP. All values represent the percent fraction of the maximum intensity.

at 20 × 20 grid points inside the laser beam cross section. It was assumed that the same intensity pattern shown in Fig. 1 is valid throughout the entire depth of field of the FSSP probe. Particles of identical size will be sized differently depending on their trajectory inside the laser beam cross section. The normalized intensity distribution values presented in Fig. 1 were used to calculate the maximum signal voltage caused by particles passing through the laser beam. Baumgardner and Spowart (1990) formula for the signal amplitude at each laser beam intensity interval was used during these calculations.

A computer code was written to estimate the distortion of size spectra by the NOAA FSSP under three different transit time modes: normal, delayed, and inhibit. For a given particle distribution, the expected size spectra by an FSSP were calculated for these three transit time modes, using the Baumgardner and Spowart's (1990) formula and the measured beam intensity profile illustrated in Fig. 1. Examples of results of such calculations are presented in Figs. 2 and 3. A true air speed of 65.0 m s^{-1}, which is typical for the NOAA King Air, was used for these calculations. The size range of the FSSP was assumed to be 2 to 32 μm, which was used for atmospheric aerosol measurements with the NOAA FSSP. A constant sample volume was assumed for all uncorrected size spectra. Effects of sample volume changes are corrected through a numerical inversion of the transformation matrix to be discussed in the following section.

Figure 2 shows how the measured size spectra would be distorted for a hypothetical constant distribution of a hundred particles per channel. It shows that the inhibit mode operation would severely overestimate the particle concentration in the lower size channels and underestimate it in the higher size channels. The normal mode operation would severely undercount the small particles while it improves its counting accuracy for large particles. The delayed mode operation shows the best results among the three spectra but a distortion of the size spectrum is still evident.

The size distributions of atmospheric aerosols are often modeled with a lognormal distribution. The result of a similar calculation is shown in Fig. 3 for a more practical lognormal distribution having a volume geometric median diameter, v_{gmd} = 15.0 μm, geometric standard deviation, σ_g = 1.75 and total volume concentration, V_c = 100 μm^3 cm^{-3}. Figure 3 shows that the distortion of the measured size spectra is noticeable only in the lower size channels for a lognormal size distribution. The relatively fewer numbers of particles available in the higher size channels resulted in insignificant differences in the measured size spectra. In order to estimate the measurement errors associated with the uncorrected FSSP data, a lognormal fit was applied to each of the three calculated size spectra in Fig. 3 using the computer fitting routine developed by Horvath et al. (1990). The volume geometric median di-

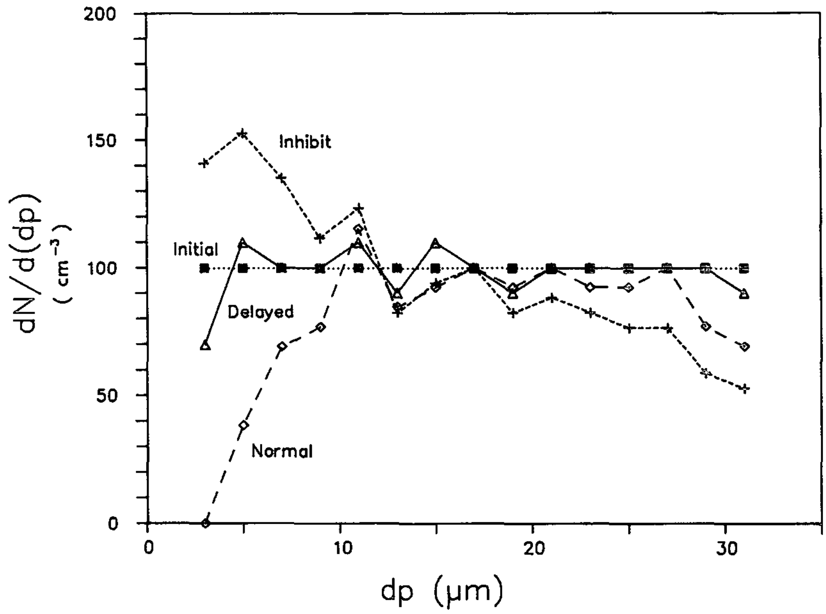

FIG. 2. Simulated size spectra of the NOAA FSSP for a uniform size distribution with a concentration of 100 particles/channel.

ameter (v_{gmd}), geometric standard deviation (σ_g), and total particle volume concentration (V_{cal}) under the fitted lognormal distributions are given in Table 1. The uncorrected FSSP size spectrum measured under the inhibit mode would underestimate the v_{gmd} as 14.30 μm and overestimate σ_g and V_{cal}; 1.78 and 115.4 μm^3 cm^{-3}, respectively. This is an overestimate of the particle volume concentration of 15.4%. The size spectrum measured under the normal mode severely underestimates the width of the distribution; σ_g = 1.65, due to the loss of counts in the first couple of size channels. The delayed mode size spectrum results in a closer fit to the initial distribution than others; but it still underestimates the volume concentration 7.1%. The same

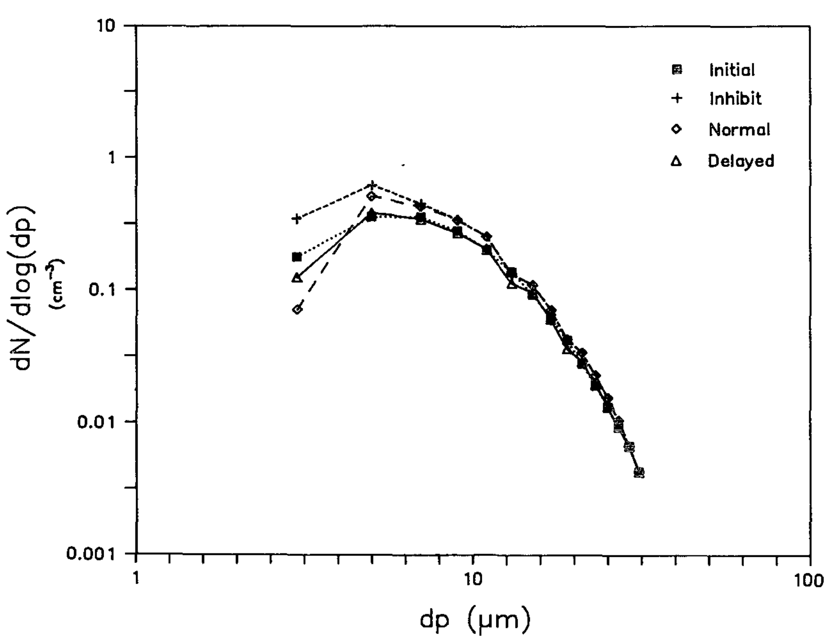

FIG. 3. Simulated size spectra of the NOAA FSSP for a lognormal distribution with v_{gmd} = 15.0 μm, σ_g = 1.75, and V_c = 100 μm^3 cm^{-3}.

calculations made for a series of lognormal distributions having different volume geometric median diameter and geometric standard deviation values resulted in similar trends as seen in Table 1.

Recent research showed that the size spectrum measured by an FSSP depends not only on optical and electronic characteristics of the instrument, but also on the shape of the ambient particle distribution (Baumgardner 1987; Baumgardner 1988). Results of Figs. 2 and 3 confirmed this conclusion and also called for a correction algorithm which can reconstruct the initial distributions from distorted size spectra, especially for atmospheric aerosol measurement applications. During the airborne aerosol measurements by the NOAA King Air, there were occasions when the on-board FSSP probe was operated under the inhibit mode. This fact along with the results in Table 1 necessitates the development of an appropriate correction algorithm for the distorted FSSP data. An iterative correction algorithm was reported which can correct the distorted FSSP spectra measured under the normal mode (Baumgardner 1988; Baumgardner and Spowart 1990). In this paper, a correction algorithm based on the non-negative least squares (NNLS) method is used to correct the FSSP spectra measured under either the inhibit or delayed mode.

3. Correction of the FSSP size spectra

The size spectrum, $\mathbf{n}(n_1, \ldots n_{15})$ measured with an FSSP can be expressed as a product of two matrices; a 15×1 column matrix, $\mathbf{n}_0$, representing the actual size distribution of ambient aerosol particles and a 15×15 transformation matrix, $\mathbf{A}$, representing the operation characteristics of the FSSP. Thus the particle count in the ith size channel of an FSSP, n_i, becomes

$$n_i = \sum_{j=1}^{15} A_{ij} n_{0j}. \qquad (1)$$

Once the transformation matrix, $\mathbf{A}_{ij}$, is known for an FSSP, the actual size distribution, $\mathbf{n}_0$ can be determined numerically by inverting Eq. (1). This type of inversion problem is common to all other optical particle counters. In this paper, the non-negative least squares (NNLS) method (Lawson and Hanson 1974) is used to reconstruct the actual size distribution $\mathbf{n}_0$ from the measured size spectrum, $\mathbf{n}$. The NNLS method had been successfully applied to similar inversion problems associated with other types of in situ laser particle counters (Holve and Self 1979; Wedding and Kim 1986).

A solution to Eq. (1) can be expressed in a matrix form as $[\mathbf{n}_0] = [\mathbf{A}]^{-1}[\mathbf{n}]$. A straight forward matrix inversion could result in negative element values in a solution vector, $[\mathbf{n}_0]$, which are physically impossible. The NNLS method finds a solution by searching for a non-negative vector $[\mathbf{n}_0]$, which minimizes the residual vector, $\mathbf{R} = [\mathbf{n}] - [\mathbf{A}][\mathbf{n}_0]$.

First, elements of the transformation matrix $[\mathbf{A}]$ were determined based on the signal pulse height calculations from the theoretical analyses by Baumgardner and Spowart (1989). Results from these calculations are summarized in Tables 2 and 3. During these calculations it was assumed that particles had an equal probability of passing through any segment of the beam cross section. Transformation matrix elements for the inhibit mode are tabulated in Table 2. It is shown that the measured size spectrum under the inhibit mode would be distorted resulting in a broader distribution than the original one. Table 3 gives the results of the transformation matrix calculations for the delayed mode. When the FSSP is operated under the 'delayed' mode, it would accept only particles passing through a narrow center region of the beam cross section, resulting in broadening of the size spectrum around only two or three size channels for a monodisperse distribution. It is difficult to apply the NNLS method for normal mode applications because the transformation matrix element values change with the shape of the ambient size distribution (Baumgardner 1988; Baumgardner and Spowart 1989).

The transformation matrix element values, A_{ij}, shown in Tables 2 and 3 are applicable for true air speeds of 65 and 100 m s^{-1}. At substantially different true air speed values, A_{ij} values should be recalculated. Tables 2 and 3 show that the degree of particle under-

TABLE 1. Lognormal fit to the FSSP size spectra.

	Initial distribution	Uncorrected			Corrected	
		Inhibit	Normal	Delayed	Inhibit	Delayed
v_{gmd}	15.0	14.30	14.35	14.85	14.90	15.14
σ_g	1.75	1.78	1.65	1.71	1.73	1.76
V_{cal}	100.0	115.4	106.0	92.9	98.4	102.5
r	—	1.000	0.998	0.999	1.000	1.000

v_{gmd}: Volume geometric median diameter (μm)
σ_g: Geometric standard deviation
V_{cal}: Particle volume concentration (μm^3 cm^{-3})
r: Correlation coefficient

TABLE 2. Transformation matrix, A_{ij} of the NOAA FSSP under inhibit mode.

	j														
i	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
							True air speed = 65 m s^{-1}								
1	0.65	0.15	0.05	0.10	0.10	0.10	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.60	0.30	0.10	0.00	0.00	0.05	0.10	0.10	0.05	0.00	0.00	0.00	0.00	0.00
3	0.00	0.00	0.45	0.20	0.10	0.10	0.00	0.00	0.00	0.05	0.10	0.10	0.05	0.00	0.00
4	0.00	0.00	0.00	0.45	0.20	0.05	0.10	0.00	0.00	0.00	0.00	0.00	0.05	0.05	0.05
5	0.00	0.00	0.00	0.00	0.45	0.20	0.10	0.10	0.10	0.00	0.00	0.00	0.00	0.05	0.05
6	0.00	0.00	0.00	0.00	0.00	0.35	0.15	0.10	0.00	0.10	0.00	0.00	0.00	0.00	0.00
7	0.00	0.00	0.00	0.00	0.00	0.05	0.35	0.15	0.15	0.00	0.10	0.00	0.00	0.00	0.00
8	0.00	0.00	0.00	0.00	0.00	0.00	0.05	0.35	0.10	0.15	0.00	0.10	0.10	0.00	0.00
9	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.05	0.30	0.10	0.15	0.00	0.00	0.10	0.00
10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.30	0.10	0.15	0.00	0.00	0.10
11	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.30	0.10	0.15	0.05	0.00
12	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.30	0.10	0.10	0.05
13	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.30	0.10	0.15
14	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.30	0.10
15	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.35
							True air speed = 100 m s^{-1}								
1	0.65	0.30	0.10	0.05	0.10	0.10	0.10	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.45	0.25	0.15	0.10	0.00	0.00	0.05	0.10	0.10	0.10	0.00	0.00	0.00	0.00
3	0.00	0.00	0.40	0.25	0.10	0.10	0.10	0.00	0.00	0.00	0.00	0.10	0.10	0.05	0.00
4	0.00	0.00	0.00	0.35	0.30	0.20	0.00	0.10	0.10	0.00	0.00	0.00	0.00	0.05	0.10
5	0.00	0.00	0.00	0.00	0.25	0.25	0.20	0.05	0.00	0.10	0.00	0.00	0.00	0.00	0.00
6	0.00	0.00	0.00	0.00	0.00	0.20	0.25	0.15	0.10	0.00	0.10	0.00	0.00	0.00	0.00
7	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.25	0.10	0.10	0.00	0.10	0.10	0.00	0.00
8	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.30	0.15	0.15	0.00	0.00	0.10	0.00
9	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.25	0.10	0.15	0.00	0.00	0.10
10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.25	0.10	0.15	0.00	0.00
11	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.25	0.10	0.15	0.05
12	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.25	0.10	0.10
13	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.25	0.10
14	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.30
15	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10

sizing deteriorates as true air speed increases from 65 to 100 m s^{-1}. Unlike the normal mode, A_{ij} values for the inhibit and delayed modes are relatively independent of the shape of the ambient size distribution. For a Gaussian laser beam, the transit time of a signal pulse is independent of its maximum amplitude for the 'delayed' mode operation (Holve 1982). The A_{ij} values for the delayed mode were calculated for different lognormal size distributions by varying the geometric mean diameter in the range 5 to 20 μm with 2.5 μm increments, and the geometric standard deviation from 1.25 to 2.0 with 0.25 increments. The A_{ij} values were found to be invariant for the delayed mode operation, which justified the use of constant A_{ij} values in the correction algorithm of the FSSP data.

A computer code (FSSPNNLS) based on the NNLS method, was written to correct the size spectra measured with an FSSP under either inhibit or delayed mode. The FSSPNNLS program was first tested for the size spectra shown in Fig. 3. After correcting the distorted size spectra in Fig. 3 using the FSSPNNLS program, the lognormal fitting routine was applied again to those corrected size spectra in order to evaluate the validity of the correction routine. Results of such calculations are included in Table 1. As shown, the correction program was able to successfully reconstruct the initial distribution well within the measurement uncertainty range for both modes. For the inhibit mode the corrected size spectrum was fitted to a lognormal distribution with v_{gmd} = 14.9 μm, σ_g = 1.73, and V_{cal} = 98.4 μm^3 cm^{-3}, resulting in satisfactory improvements in reconstructing the original distribution. The correction program also improved the accuracy of recovering the original distribution for the delayed mode data.

Similar analyses were performed for a series of different lognormal distributions having a combination of different geometric mass median diameter and geometric standard deviation values; v_{gmd} = 5.0, 7.5, 10.0, 12.5, 15.0, 17.5 and 20.0 μm, and σ_g = 1.25, 1.5, 1.75 and 2.0. The FSSPNNLS program succeeded in reconstructing the initial distributions for all cases. Thus, it can be concluded that the correction algorithm developed in this study, based on the NNLS method, is appropriate for reconstructing the original aerosol size distributions from the distorted size spectra measured with an FSSP under the inhibit or delayed mode.

4. Applications of the correction algorithm

Theoretical validation of the FSSP data correction algorithm developed in this paper was discussed in the preceding section. However, experimental validation of this correction algorithm is not easy because standard calibration particles of known optical properties and size distribution, preferably a lognormal distribution similar to atmospheric aerosols of interest, are not available. During a field experiment in February 1989 over New Mexico, the NOAA King Air aircraft flew in preliminary test flight on 7 February 1989. Aerosol size distributions were measured with the NOAA FSSP along a horizontal flight path for a short period of time lasting 1-min each under inhibit and delayed modes. The total sampling time in each mode was kept as short as possible in order to collect similar samples during the two sampling modes, inhibit and delayed.

Results of lognormal fits to these test flight data are summarized in Table 4. A lognormal fit to the uncorrected FSSP data resulted in wide discrepancies between the fitted distributions, based on inhibit and delayed mode size spectra. The uncorrected inhibit mode data resulted in a smaller mean diameter ($v_{gmd} = 5.89$ μm) and a larger aerosol volume concentration ($V_{cal} = 7.82\ \mu m^3\ cm^{-3}$) than the delayed mode data; $v_{gmd} = 6.31$ μm and $v_{cal} = 5.90\ \mu m^3\ cm^{-3}$, respectively. After correcting the measured FSSP data with the FSSPNNLS program, the two fitted distributions agreed reasonably well with each other as shown in Table 4. We conclude that the two corrected size distributions represent essentially the same sample of aerosol particles although the two uncorrected distributions might suggest otherwise. The test results shown in Table 4 are based on a rather limited dataset. More test measurements need to be taken under different atmospheric conditions. Such test flights are planned in the near future. Nevertheless, the results shown in Table 4 confirmed the necessity and validity of the correction algorithm for FSSP data, especially those measured under the inhibit mode.

The correction algorithm was also applied to a set of FSSP data measured for marine aerosols. All FSSP

TABLE 3. Transformation matrix, A_{ij} of the NOAA FSSP under delayed mode.

	j														
i	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
	True air speed = 65 m s^{-1}														
1	0.35	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.50	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	0.00	0.00	0.45	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.00	0.45	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5	0.00	0.00	0.00	0.00	0.45	0.10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
6	0.00	0.00	0.00	0.00	0.00	0.35	0.10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
7	0.00	0.00	0.00	0.00	0.00	0.05	0.35	0.10	0.05	0.00	0.00	0.00	0.00	0.00	0.00
8	0.00	0.00	0.00	0.00	0.00	0.00	0.05	0.35	0.05	0.05	0.00	0.00	0.00	0.00	0.00
9	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.05	0.30	0.05	0.05	0.00	0.00	0.00	0.00
10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.30	0.05	0.05	0.00	0.00	0.00
11	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.30	0.05	0.05	0.00	0.00
12	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.30	0.05	0.05	0.00
13	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.30	0.05	0.05
14	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.30	0.10
15	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10	0.35
	True air speed = 100 m s^{-1}														
1	0.30	0.20	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
2	0.00	0.45	0.25	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	0.00	0.00	0.40	0.25	0.10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.00	0.35	0.30	0.20	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
5	0.00	0.00	0.00	0.00	0.25	0.25	0.20	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.00
6	0.00	0.00	0.00	0.00	0.00	0.20	0.25	0.15	0.10	0.00	0.00	0.00	0.00	0.00	0.00
7	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.25	0.10	0.10	0.00	0.00	0.00	0.00	0.00
8	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.20	0.30	0.15	0.15	0.00	0.00	0.00	0.00
9	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.25	0.10	0.15	0.00	0.00	0.00
10	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.25	0.10	0.15	0.00	0.00
11	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.25	0.10	0.15	0.05
12	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.25	0.10	0.10
13	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.25	0.10
14	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.15	0.30
15	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.10

TABLE 4. Lognormal fit to the test flight data of 7 February 1989.

	Uncorrected		Corrected	
	Inhibit	Delayed	Inhibit	Delayed
v_{gmd}	5.89	6.31	6.39	6.20
σ_g	1.60	1.58	1.60	1.70
V_{cal}	7.82	5.90	4.94	5.02
r	0.987	0.989	0.993	0.991

v_{gmd}: Volume geometric median diameter (μm)
σ_g: Geometric standard deviation
V_{cal}: Particle volume concentration (μm^3 cm^{-3})
r: Correlation coefficient

data measured during the 1988 Coordinated Air-Sea-Western Atlantic Ocean Experiment (CASE-WATOX) program (Boatman et al. 1990) were collected under the inhibit mode by the NOAA FSSP. Consequently, all the measured FSSP size spectra were corrected using the FSSPNNLS program and were fitted to lognormal distributions. One example of results from such correction calculations is summarized in Table 5. Figures 4 and 5 show the uncorrected and corrected size spectra measured with the FSSP under the inhibit mode in the free troposphere and in the marine boundary layer, respectively. It shows that the uncorrected FSSP data underestimated the volume median diameter by 6.7% and 9.3%, and overestimated the volume concentration by 49.2% and 50.2% for the free troposphere and marine boundary layer data, respectively. It is shown that the uncorrected FSSP data resulted in overestimation of the aerosol volume concentration by as much as about 50%. Thus, it is imperative to correct the FSSP data measured under the inhibit mode, considering the effects of particle trajectory and beam intensity profile inside the laser beam cross section.

5. Discussion and conclusions

The effects of particle trajectory and beam intensity profile on the size spectra measured with the NOAA FSSP under different transit time modes have been investigated through theoretical calculations based on the analyses by Baumgardner and Spowart (1990). It was shown from these calculations that the size spectra measured by the FSSP is distorted, depending on various parameters such as the response time and beam intensity profile of the probe, sampling speed, shape of the size distribution, and the choice of transit time mode. A correction algorithm is reported in this paper to reconstruct the original distributions from the distorted size spectra measured under inhibit or delayed mode using a numerical inversion scheme based on the non-negative least squares (NNLS) method. This correction algorithm was confirmed to be able to recover the initial lognormal distributions from the simulated size spectra. The same correction algorithm was also applied to experimentally measured FSSP size spectra of background atmospheric aerosols. It revealed that this correction procedure was necessary in order to accurately estimate the aerosol size distribution of atmospheric aerosols with an FSSP.

TABLE 5. Lognormal fit to 1988 CASE-WATOX marine aerosol data measured on 17 July 1988 at 2590 and 150 m MSL.

	Free troposphere (2590 m MSL)		Marine boundary layer (150 m MSL)	
	Uncorrected	Corrected	Uncorrected	Corrected
v_{gmd}	6.08	6.52	6.21	6.85
σ_g	1.69	1.71	1.89	1.88
V_{cal}	2.64	1.77	63.4	42.2
r	0.998	0.998	0.998	0.998

v_{gmd}: Volume geometric median diameter (μm)
σ_g: Geometric standard deviation
V_{cal}: Particle volume concentration (μm^3 cm^{-3})
r: Correlation coefficient

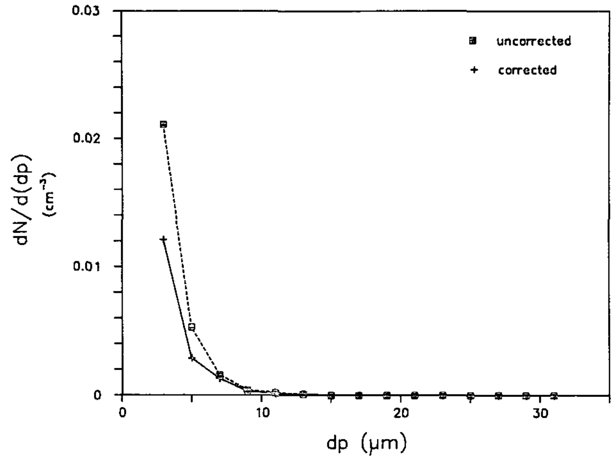

FIG. 4. Uncorrected and corrected size spectra measured with an FSSP in the free troposphere on 17 July 1988 during the CASE/WATOX program.

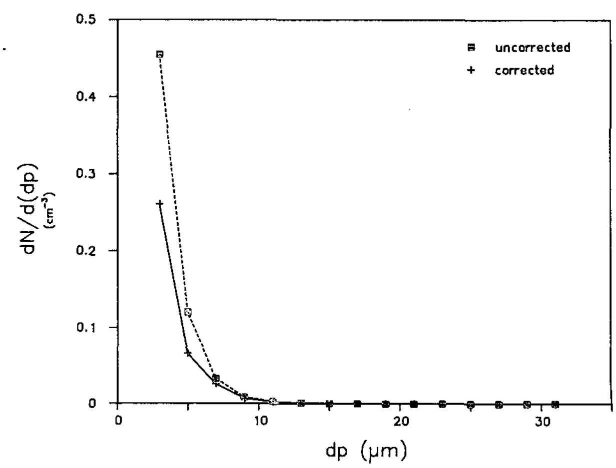

FIG. 5. As in Fig. 4 but in the marine boundary layer.

A new version of the FSSP probe (FSSP 300) has recently been introduced by Particle Measuring Systems Inc., which uses a masked slit instead of velocity reject circuitry to reduce the effects of particle trajectory on particle sizing. This type of instrument would suffer similar measurement problems encountered with the current FSSP under the 'inhibit' mode operation because of electronic response time limitations and nonuniform laser beam intensity distribution. Thus, the correction method developed in this study could be readily adopted for analyzing size spectra data measured with an FSSP-300 probe.

In this study, the size calibration provided by the manufacturer was assumed to be correct while correcting the shape of size spectra measured with an FSSP. However, particle sizing error is unavoidable with an FSSP when the optical properties of the measured particles are different from those of calibration particles, which are water droplets. Kim and Boatman (1990) reported new size calibration data as a function of aerosol type and relative humidity for atmospheric aerosol measurement applications. In order to complete the accurate estimation of aerosol size distribution, correct size calibration data would have to be used, based on the optical properties of the measured particles, after correcting the shape of the size spectra using the correction algorithm developed in this study.

Another important correction associated with an FSSP is the effect of optical coincidence and electronic dead time loss. They are main sources of errors when an FSSP is used to measure cloud droplet spectra with high number concentrations. Cooper (1988) reported that the problem of coincidence became serious in clouds having droplet concentrations over 500 cm^{-3}. For cloud droplet spectra measurements, corrections for coincidence should be made in addition to the corrections of size spectra discussed in this study. However, coincidence error is insignificant for atmospheric aerosol measurements where particle number concentration are usually less than 10 cm^{-3}.

It has been known to users of the FSSP instrument that the size spectra measured with an FSSP might be distorted due to inherent optical and electronic limitations of the instrument. The FSSPNNLS correction scheme reported in this paper would recover more closely the real size distributions of atmospheric aerosols from the FSSP data measured under 'inhibit' or 'delayed' mode. Such correction to the FSSP data can yield a much better understanding of the microphysical structure of cloud droplets as well as atmospheric aerosols.

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