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AN APPROACH TO EVALUATING AND CORRECTING AERODYNAMIC PARTICLE SIZER MEASUREMENTS FOR PHANTOM PARTICLE COUNT CREATION

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An Approach to Evaluating and Correcting Aerodynamic Particle Sizer Measurements for Phantom Particle Count Creation*

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An aerodynamic particle sizer (APS) can be used to make real-time measurements of the aerodynamic particle size distribution over the range of 0.5 to 32 μm. This instrument is very useful in conducting health-related aerosol measurements involving aerosol generation, respirator efficiency, and particulate sampling efficiency. One of the two signal processors within the APS can create spurious or phantom particle counts that can significantly affect relative measurements and calculated mass distributions. In the APS, particle size measurement is based upon a particle's transit time between two laser beams that are perpendicular to an accelerating airflow. The signal processors measure each particle's transit from the time between the two pulses of scattered light that are generated as the particle passes through the two laser beams. When only a single pulse from a particle is detected, another pulse can cause the recording of a randomly sized phantom particle. The small particle processor (SPP), which measures particle transit from the times in digital increments of 4 nanoseconds, can create phantom particles; the large particle processor (LPP), which measures particle transit times in digital increments of 66.67 nanoseconds, is designed to prevent the creation of phantom particles. These two processors overlap in the range of 5.2 to 15.4 μm. The difference in particle counts in this overlap region can be used to estimate an upper limit to the number of phantom particles in each channel of data from the SPP. The user needs to exercise some caution in using this correction because the LPP of the APS responds to coincidence by underestimating the particulate concentration. Furthermore, this correction does not cleanse the data of the statistical noise caused by phantom particle creation.

Time-of-flight (TOF) aerosol spectrometers are used to make real-time measurements of an aerodynamic particle size distribution. An example of such an instrument is the aerodynamic particle sizer (APS, Model 3310, TSI Inc., St. Paul, Minn.), which measures approximate aerodynamic diameter over the range of 0.5 to 32 μm. This instrument measures aerosol concentration as an approximate function of aerodynamic diameter, i.e., the diameter of a unit density sphere that has the same settling velocity in still air as the particle in question. Because aerosol behavior, especially in the human respiratory system, varies with aerodynamic particle diameter, such instruments are very useful in conducting industrial hygiene–related aerosol research.[1–3] Baron noted that excess background counts were created by the APS.[4] While using the APS to study filter penetration, Wake concluded that the APS can generate phantom or spurious particles.[5] Heitbrink et al.[6] developed expressions for estimating the concentration of phantom particles created by the APS. The purpose of this paper is to present and discuss a method for correcting the APS data for phantom particle creation.

In order to understand phantom particle creation, the measurement techniques used in the APS need to be understood. Instead of directly measuring settling velocity, particle size measurement in the APS is based on the principle that the magnitude of a particle's velocity in an accelerating airflow is closely related to the particle's aerodynamic diameter. To measure the particle velocity, the APS measures the time difference between the two electronic pulses that are generated as each particle passes through the two laser beams positioned just below the acceleration nozzle.[7]

Generally, the transit time in the APS is related to aerodynamic diameter through calibration with monodisperse spheres of known density (e.g., polystyrene latex). Because the APS measures particle transit times in a high-velocity, nonStokesian flow regime, the measured aerodynamic diameter is corrected for the effects of gas viscosity and pressure and the particle's shape and density.[8,9] Except for extreme cases of high or low density or large shape factors, the corrections are typically on the order of 20%.

Figure 1 illustrates how the APS can interpret pulses to create nonexistent particles (phantom particles). The first particle's pulses are A_1 and A_2; the second particle's pulses are B_1 and B_2. The second pulse (A_2) of the first particle is too small to be detected. Single pulses can occur because of noise, particle

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asymmetry, or differences in beam intensity. The electronic timer stays on for a time (T_{max}) that defines the largest particle that can be sized. If a second particle arrives before the timer is turned off and the first pulse (B_1) of that particle is detected, the timer is terminated and the time between pulses A_1 and B_1 is recorded as a transit time. As a result, two real particles are deleted and one phantom particle is added to the measured size distribution. The latter has been defined as an open-timer phantom particle.[6] Pulse B_2 starts the timer again, but it is ignored unless another pulse is detected before time T_{max} elapses. Because the arrival time of the second particle is a random event, the phantom particles can occur anywhere in the measured size range.

In order to correct the APS data for phantom particle creation, the details of the instrument's detection system and data manipulation need to be considered. The APS uses a single photomultiplier tube to detect the two pulses of light scattered by a particle as it passes through the two laser beams. Two signal processors, the small particle processor (SPP) and the large particle processor (LPP), independently use the time difference between the detector pulses to measure a particle's transit time and, hence, particle size. As illustrated in Figure 2, the SPP and LPP provide particle size information over two overlapping size ranges, resulting in a manufacturer-specified total detection range of 0.5 to 32.8 μm. When the software supplied with the instrument is used, size distribution data for particle sizes smaller than 5.233 μm (the lower boundary for Channel 36) are obtained just from the SPP aerodynamic size channels. For particles larger than 15.399 μm (the lower boundary for Channel 51), particle size data are obtained just from the LPP aerodynamic size channels. As particle size increases from 5.233 to 15.399 μm (Channels 36 to 50), an arbitrary weighing scheme is used to phase out the SPP size data and to phase in the LPP data.[10]

The SPP and LPP record the number of particles counted in equal increments of transit time, which are called accumulator channels. The calibration curve for the APS relates particle size to accumulator channel. A printed listing of the maximum particle diameter for a channel and its accumulator channel can be generated during the calibration procedure. The number of counts in each particle size channel is obtained by summing the counts in the appropriate accumulator channels.

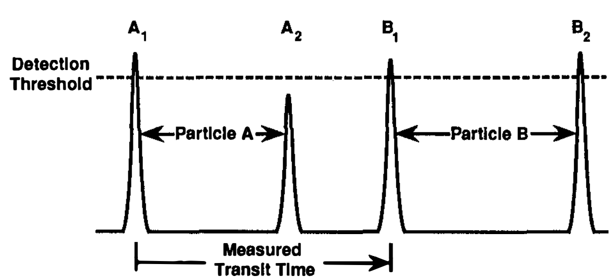

FIGURE 1. Schematic of processes for creating phantom particles by the APS. Pulse A_2 of Particle A is not detected. As a result, the time between Pulse A_1 and Pulse B_1 is measured as a transit time.

In order to measure aerodynamic diameter, the SPP measures each particle's transit times digitally in increments of 4 nanoseconds (nsec) from 750 to 4096 nsec and stores the counts in Accumulator Channels 128 to 1024. The particle counting rules for the SPP are listed below.

1. When a pulse larger than 0.02 V is detected, the timer is started.
2. If the first pulse is larger than 0.75 V, the threshold for the second pulse is increased from 0.02 to 0.75 V.
3. After the first pulse has been detected, all following pulses are masked for a 750-nsec dead-time period (the transit time for air).
4. If a second pulse is detected before 4096 nsec have elapsed, a particle is counted and sized and the timer is reset to zero.
5. If 4096 nsec elapse after the first pulse and no other pulses have been detected, the timer is reset to zero.
6. While the timer is being reset, no pulses can be sensed for a period of 200 nsec.

As indicated in Figure 1, an open-timer phantom particle is created when a single pulse is followed by either another single pulse or a fully detected particle arrives between 750 and 4096

	A	B	C	D	E	F
1	MAXIMUM	Midpoint	Channel	Initial	$N_{ph,i}$	Corrected
2	ACCUMULATOR	Diameter	Number	File		File
3	CHANNEL	(μm)		(N_i)		($N_{i,adj}$)
4	195.36					
5	196.14		0	57	2.48	54.5
6	196.96		1	65	2.61	62.4
7	197.86	<.486	2	82	2.87	79.1
8	198.88	0.504	3	129	3.25	125.8
.	SPP .	.	.	.	.	.
.	.	.	.	.	.	.
40	408.01	5.04	35	428	38.2	389.8
41	422.00	5.42	36	440	44.5	395.5
.	SPP IN .	.	.	.	.	.
.	OVERLAP .	.	.	.	.	.
.	REGION .	.	.	.	.	.
53	691.68	12.8	48	1173	92.12	1080.9
52	721.69	13.8	49	1078	95.59	982.4
55	752.77	14.8	50	951	99.00	852.0
56	784.95	15.9	51	721	.	721.0
.	.	.	.	.	.	.
.	.	.	.	.	.	.
.	LPP .	.	.	.	.	.
.	.	.	.	.	.	.
66	1290.18	32.8	61	1	.	1.0
67	1371.72	35.2	62	1	.	1.0
68	.	5.42	63	38	.	395.5
69	.	5.82	64	58	.	442.6
.	LPP IN .	.	.	.	.	.
.	OVERLAP .	.	.	.	.	.
.	REGION .	.	.	.	.	.
.	.	.	.	.	.	.
82		14.8	77	852	.	852.0
83		15.9	78	721	.	721.0
84			79	"N"	.	"N"
85						
86	N_{ph} =	3.18				
87						
88	the first line of the aps data file:					
89	15,0,1,0,"","07-06-1989 SN 301"					

FIGURE 2. An abbreviated spreadsheet that illustrates the adjustment of APS data for phantom particle creation. The entry in Line 89, Column A is the first line of the APS data file. The letters and numbers in gray define the columns and rows in the spreadsheet.

nsec after the first pulse. The concentration of open-timer phantom particles, C_{ot}, recorded by the SPP is[6]

$$C_{ot} = C_{sp}[(C_d + C_{sp})QT] \quad (1)$$

C_d = concentration of particles that have been completely detected by the APS

C_{sp} = concentration of single pulses

Q = sampling rate of APS inner nozzle (16.6 cm^3/sec)

T = DEF = time period for sensing second pulse (3.3 µsec)

The term within the square brackets on the right-hand side of Equation 1 is the expected number of counts per single pulse that can close the timer before it resets. At low concentrations, this term approximates the probability of another single pulse or fully detected particle closing the timer. This expression assumes that open-timer phantom particle creation occurs because of coincidence between a single pulse and either a completely detected particle or another single pulse. The expression ignores the possibility of coincidence among three or more particles. Because the arrival time of the second pulse is a random event, the open-timer phantom particles approximately follow a random uniform distribution with respect to transit time.[6] Thus, the expected number of phantom particles per accumulator channel is constant. Minor deviations from this distribution occur because of processor dead time (Item 3 of the SPP particle counting rules).

The large particle processor (LPP) is designed to eliminate coincidence-produced phantom particles from the measured size distribution. The LPP measures particle transit times in 66.67 nsec increments and it stores these counts in Accumulator Channels 0 to 127. The counting rules for the LPP are listed below.

1. Each pulse in the pulse pair must have an amplitude greater than 3 V.
2. No pulse greater than an interference threshold of 0.5 V may occur less than 8400 nsec before the first pulse of the pulse pair, within 8400 nsec of the trailing edge of the second pulse, or between the two pulses.
3. The maximum transit time is 8400 nsec.
4. When the timer is being reset, all incoming pulses are ignored for an 840-nsec period. The timer is reset when extraneous pulses are detected, when the time exceeds maximum transit time, and when a particle is registered by the LPP.

In order for the LPP to count a particle, the pulses must be completely isolated from other potentially interfering pulses. If a second particle with a pulse amplitude greater than 0.5 V arrives within 8400 nsec of the first particle, both particles will be rejected by the LPP. This logic causes the rejection of all particles whose pulse pairs overlap, as well as some particles whose pulse pairs do not overlap and might have been properly sized.

CORRECTION FOR PHANTOM PARTICLE CREATION

Because it is nearly impossible for the LPP to create phantom particles, the difference in counts measured by the SPP and LPP at the largest particle size reported by the SPP can be used to estimate the number of phantom particles counted (Annotation B in Figure 3). From a calibration printout supplied by the APS software, the transit time width (as the number of accumulator channels) of the largest SPP particle size channel can be obtained. This allows estimation of the number of phantom particles per accumulator channel:

$$N_{ph} = \frac{N_{50} - N_{77}}{\Delta A_{50}} \quad (2)$$

ΔA_{50} = transit time width in terms of the accumulator channels for the last SPP channel, which is Channel 50

N_{50} = number of particles counted in the largest SPP particle size

N_{77} = number of particles counted in the largest LPP overlap channel

Assuming that there are no coincidence losses by the LPP in N_{77}, the number of phantom particles in each channel ($N_{ph,i}$) and the adjusted number ($N_{i,adj}$) of counts in each SPP channel can be computed as follows.

$$N_{ph,i} = \Delta A_i \, N_{ph} \quad (3)$$

$$N_{i,adj} = N_i - N_{ph,i} \quad (4)$$

ΔA_i = accumulator channel increment for SPP Channel i

N_i = number of particles counted in SPP Channel i

As illustrated in Figure 2, the adjustment of the data can be performed by using a spreadsheet program. The data collected by the APS are stored as ASCII text files that can be imported into the spreadsheet (Column D). The particle channel number, the particle diameter, and the accumulator channel

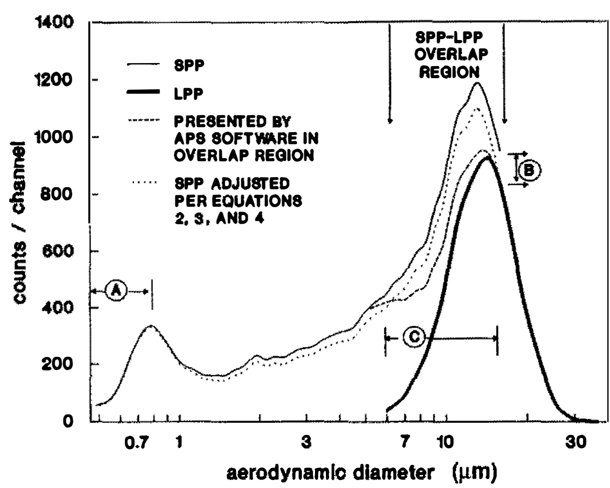

FIGURE 3. The correction of APS data for phantom particle creation. The annotations illustrate the correction of the SPP data for phantom particle creation: (A) partially detected particles cause phantom particle creation; (B) the largest overlapping channel is used to correct the SPP data for phantom particle creation; and (C) because the LPP does not efficiently detect smaller particles, only the largest overlapping channel was used in this correction.

(which is obtained from the calibration printout) are entered into the spreadsheet (Columns A, B, and C, respectively). By using Equations 2, 3, and 4, the estimated number of phantom particles in each SPP channel is computed (Column E) and the adjusted number of SPP counts is computed (Column F). In the LPP overlap region of Column F, LPP counts are replaced by the adjusted SPP counts. The file header (Cell A89) and the adjusted data (Cells F5–F83) can be assembled into an adjusted APS data file. In Lotus 1-2-3® (Version 2.1, Lotus Development Corp, Cambridge, Mass.), this can be accomplished by specifying the "file" and "unformatted" options when using the print command.

In Figure 3, the correction of APS measurements for phantom particle creation is annotated to facilitate discussion of the correction procedure. Incomplete detection of particles smaller than 1 µm (Annotation A in Figure 3) causes the creation of phantom particles, resulting in a positive bias in the measurements made by the SPP. The difference between the SPP and the LPP at the largest overlapping particle size channel (Annotation B in Figure 3) is used to adjust the SPP data for phantom particle creation. Also note that the magnitude of this adjustment increases with increasing particle size. The last overlapping channel is used for this correction because the LPP does not appear to efficiently detect particles smaller than 10 µm (Annotation C in Figure 3).

When the pulse amplitudes were measured by using a storage oscilloscope (Model 7623A, Textronix, Beaverton, Oreg.) with the procedures described in the APS manual, the pulse amplitudes for particles in the 5–8 µm range were between 1.2 and 2 V. For particles in the 12–16 µm range, pulse amplitudes were between 3 and 5 V. Thus, as particle size increases from 5 to 16 µm, the pulse amplitude rises above the 3-V pulse detection threshold of the LPP. The manner in which the data from the two processors are blended by the presentation software can obscure any detection threshold effect associated with the LPP. For comparison, Figure 3 also presents the blended distribution in the overlap region as calculated by the current APS software.[10]

ASSUMPTIONS IN USE OF CORRECTION FORMULAS

Using Equations 2 and 3 to correct the SPP data for phantom particle corrections involves two assumptions.

1. The detection efficiency for the SPP and the LPP are both 100% at the largest overlap channel.
2. The SPP is forming phantom particles because of coincidence between a single pulse and either another single pulse or a fully detected particle.

Because the LPP has a much higher pulse detection threshold than the SPP, the LPP may count fewer real particles than the SPP. In addition to losing particles to inadequate pulse amplitude, the LPP deletes particles because of coincidence. Because Equations 2, 3, and 4 ignore LPP detection losses, $N_{ph,i}$ represents an upper limit to the number of phantom particles in each channel.

The above considerations are based upon coincidence between two particles. As long as the total concentration of fully detected particles and single pulses sensed by the SPP remains below 1000 (particles and single pulses)/cm^3, the error in $N_{ph,i}$ caused by multiple particle coincidence will be less than 5%.

STATISTICAL CONSEQUENCES OF CORRECTION

Although much of the bias associated with phantom particle creation can be removed, the statistical variability associated with phantom particle creation will remain in the corrected data. To be sure that real particle counts have been recorded in each channel, the limits of detection and quantitation that are used for reporting the results of environmental analysis can be applied.[11] The standard deviation, $s_{i,adj}$, for the adjusted number of counts in the i-th SPP channel can be computed by pooling the standard deviation for the total number, N_i, particles counted and the standard deviation for estimated number, $N_{ph,i}$, of phantom particles in each channel:

$$S_{i,adj} = \sqrt{N_i + N_{ph,i}\frac{\Delta A_i}{\Delta A_{ph}}} \quad (5)$$

At the limit of detection, which is 3 $s_{i,adj}$, there is a 99% probability that the adjusted count is above the noise. At the limit of quantitation, which is 10 $s_{i,adj}$, the uncertainty in the adjusted counts is ±30% at the 99% level of confidence. Figure 4 shows the result of adjusting the data for phantom particle creation and how the limits of detection and quantitation vary with particle size for an experiment that was described elsewhere.[6]

The data taken in Figure 4 were collected by drawing air through a sampling train, which eliminated particles larger than 10 µm.[6] Thus, all of the particles larger than 10 µm are phantom particles. For particles larger than 10 µm, the number of counts in the adjusted

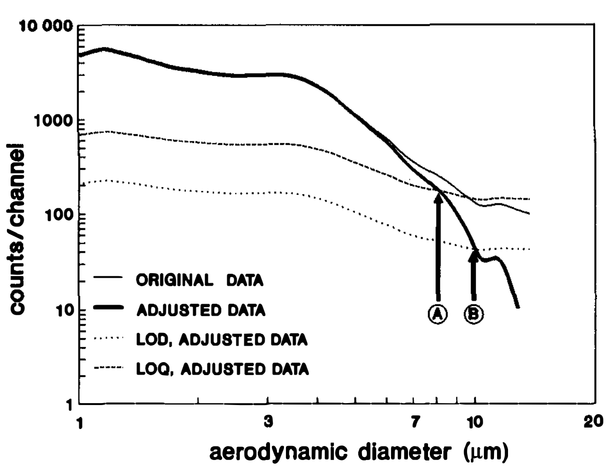

FIGURE 4. Application of the limits of detection and quantitation to APS SPP data obtained by sampling the aerosol downstream of an impactor and a settling tube, which combined to eliminate particles larger than 10 µm. The measured total concentration and the single pulse concentration for the SPP were 316 particles/cm^3 and 240 pulses/cm^3, respectively. As particle size increases between Points A and B, the measured distribution becomes less distinguishable from the statistical noise.

SPP data are below the limit of detection for the adjusted distribution. If the corrections described in this paper were perfect, the adjusted number of counts in a channel would be fluctuating around zero for particles larger than 10 μm and the number of counts per particle size channel would be increasing for particles larger than 10 μm. Instead, as particle size increases above 10 μm, the number of counts in the original distribution is constant and the number of counts in the adjusted distribution is decreasing toward zero. This behavior is probably caused by coincidence between a single pulse and a fully detected particle arriving during the detector's dead time. The number of phantom counts per accumulator channel is constant for transit times, which are greater than the sum of the transit time for a 10-μm diameter particle and the 750-nsec dead time. When a single pulse triggers the timer and a fully detected particle arrives during the dead-time period, the second pulse from the fully detected particle can close the timer. Apparently, the presence of these dead-time phantom particles caused a small, but noticeable, overestimation of the single pulse concentration and the number of phantom particles in each channel.

DISCUSSION AND CONCLUSIONS

Phantom particle creation causes the SPP to overcount the number of particles in a channel. The current APS presentation software deals with this bias by presenting a linear combination of the SPP and LPP particle counts in the overlap region. In this region, the LPP tends to undercount because of coincidence and inefficient particle detection (especially highly light-absorbing particles). Thus, the technique used in the presentation software combines two biased measurements in the hope that the average is correct.

An alternate approach is to use Equations 2, 3, and 4 to correct the SPP data for phantom particle creation. Once corrected, the final distribution can be formed by combining the adjusted SPP data and LPP data for the particles that are too large to be sized by the SPP. Finally, the limits of quantitation or detection can be used to evaluate the utility of the data for a particular application. In Figure 4, the distribution data can be used confidently up to Particle Size A and with less confidence up to Particle Size B.

When adjusting APS data by using the approach described in this paper, the undercount by the LPP needs to be evaluated. Coincidence losses by the LPP cannot be directly evaluated because the concentration of interfering particles (particles with pulses larger than 0.5 V) is not measured by the APS. Upper and lower limits can be placed on the LPP coincidence losses by using, respectively, the total aerosol concentration and LPP-measured aerosol concentration as the actual concentration, C_a, in the following formula[12]:

$$C_i = C_a e^{-QTC_a} \qquad (6)$$

C_i = the indicated concentration (particles/cm^3)

Q = the flow rate of the APS (16.67 cm^3/sec)

T = maximum time period for sensing an interfering pulse (25.2×10^{-6} sec = the sum of the maximum transit time measured by the LPP and the two 8.4-μsec time intervals in Item 2 of the LPP counting rules)

LPP undercounts because of coincidence can be kept below 4% by keeping the total aerosol concentration measured by the APS below 100 particles/cm^3. Inadequate pulse amplitude can also cause the LPP to undercount particles. At very low concentrations where phantom particle creation should not occur, the ratio of LPP to SPP counts can be used as a measure of the size-dependent detection efficiency of the LPP. In addition, the analog output of the APS and a storage oscilloscope can be used to evaluate whether a particle's pulses are large enough to be detected by the LPP.

If the processors of the APS were modified to count the number of single pulses (the number of times a processor was reset without adding a particle to the measured distribution), Equation 1 could be used to estimate the adjustment for phantom particle coincidence. This would eliminate the need to use the procedures described in this paper.

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