Ambient aerosol is known to affect global climate and reduce atmospheric visibility\textsuperscript{[1]}. Recently, epidemiological studies have also shown an association between aerosol mass concentration and increased morbidity and mortality rates\textsuperscript{[2]}. Therefore, how to measure aerosol mass concentration is a focus of ongoing research. Various methods have been used for aerosol mass concentration measurement. The light scattering method is the most popular one, because it is fast, nonintrusive and suitable for on-line measurement. It includes an ensemble-measuring technique and a single particle counting technique. The former based on a simultaneous occurrence of an ensemble of particles in sensing volume can only measure mass concentration, and has been widely employed\textsuperscript{[3−5]}. The latter based on the scattering of a single particle can simultaneously measure number and mass concentration. However, studies focusing specifically on the single particle counting technique for mass concentration measurement are few. In this letter, a model for measuring aerosol mass concentration based on the two-step calibration method is proposed. The experimental results demonstrate that mass concentrations calculated by the model are in good agreement with those measured by a norm-referenced instrument. For samples of soot and air, the slopes of fitting lines of data points are 0.9582 and 0.9220, and the correlation coefficients are 0.9991 and 0.9965, respectively.

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A model for measuring aerosol mass concentration by an optical particle counter is presented using the conception of the average mass. In this model, to understand the meaning of the pulse height distribution of particles which is used to inverse mass concentration, the relationship among intensity distribution in the optical sensing volume, particle shape, and the pulse height distribution is discussed. To solve the instability of the equivalent factor, a novel two-step calibration method is proposed. The experimental results demonstrate that mass concentrations calculated by the model are in good agreement with those measured by a norm-referenced instrument. For samples of soot and air, the slopes of fitting lines of data points are 0.9582 and 0.9220, and the correlation coefficients are 0.9991 and 0.9965, respectively.

Aerosol mass concentration is the total mass of particles in per unit volume. In order to obtain the model for measuring aerosol mass concentration using an OPC, according to the Mie scattering theory, the calculation formula for mass concentration of spherical particles under the condition of the uniform intensity distribution in optical sensing volume was derived as\textsuperscript{[6]}

\[
C_a = k_s \sum_i s(v_i) v_i^{1.5},
\]

where \( k_s \) is the proportional coefficient, which has the same dimension with mass concentration, \( v_i \) is the relative median voltage in the \( i \)th channel of an OPC, \( s(v_i) \) represents the number of voltage pulses (spherical particles) in the \( i \)th channel in a measurement cycle. Furthermore, to calculate mass concentration of irregularly natural particles, a theoretical formula has been derived to analyze the feasibility of measurement of aerosol mass concentration with an OPC\textsuperscript{[7]}:

\[
C_{th} = \int_{D_F \text{ min}}^{D_F \text{ max}} h(D_F) \overline{M}(D_F) dD_F,
\]

where \( D_F \text{ max} \) and \( D_F \text{ min} \) are the maximal and minimal optical equivalent diameters of particles, respectively; \( \overline{M}(D_F) \) is the average mass of particles with optical equivalent diameter \( D_F \); \( h(D_F) \) is the optical equivalent size distribution which represents the number of particles with optical equivalent diameter \( D_F \) in per unit volume:

\[
h(D_F) = \int_{D_V \text{ min}(D_F)}^{D_V \text{ max}(D_F)} L(D_V, D_F) dD_V,
\]
where $D_{V\text{max}}(D_F)$ and $D_{V\text{min}}(D_F)$ are the maximal and minimal volume equivalent diameters of particles with optical equivalent diameter $D_F$, respectively; $L(D_V,D_F)$ is the two-dimensional (2D) particle size distribution of non-spherical particles. From Eq. (2), we can come to a conclusion that aerosol mass concentration can be obtained with an OPC if $h(D_F)$ and $\overline{M}(D_F)$ are available. In the following we will analyze them in detail.

In our measurement system, the light source is a semiconductor laser (650 nm, 5 mV), and the relative intensity distribution of a central section in the optical sensing volume is shown in Fig. 2. Obviously, the intensity distribution is nonuniform. Thus, taking the nonuniform intensity distribution and irregularly shaped particles into account, the optical equivalent size distribution of particles $h(D_F)$ will be discussed.

To reduce complexity of the problem, two probabilities are introduced. 1) Assuming that particles are spherical and intensity distribution is nonuniform. Scattered light flux of a spherical particle is usually different when it passes through different places in the optical sensing volume, making the particles possess many optical equivalent diameters. Under this condition, if optical equivalent diameter of the particle $D_{F_s} \in [D_{F\text{min}}, D_{F\text{max}}]$, and then $p_s(D_{F_s}|D_V)$ represents the probability that the spherical particle with diameter $D_V$ possesses the optical equivalent diameter $D_{F_s}$. 2) Assuming particles are irregular and intensity distribution is uniform. Scattered light flux of an irregular particle is different when spatial orientation of the particle in the optical sensing volume is different, making the particles possess many optical equivalent diameters. In this case, if optical equivalent diameter of the particle $D_{F_s} \in [D_{F\text{min}}, D_{F\text{max}}]$, and then $p_o(D_F|D_V)$ represents the probability that the irregular particle with volume equivalent diameter $D_V$ possesses the optical equivalent diameter $D_{F_o}$.

Combining the two points mentioned above, in the case of nonuniform intensity distribution and irregular particles, assuming optical equivalent diameter of a particle $D_F \in [D_{F\text{min}}, D_{F\text{max}}]$, and then $p(D_F|D_V)$ represents the probability that the particle with volume equivalent diameter $D_V$ possesses the optical equivalent diameter $D_{F_o}$. Due to the independence of intensity distribution in the optical sensing volume and particle shape, we have

$$p(D_F|D_V) = p_s(D_{F_s}|D_V)p_o(D_{F_o}|D_V). \quad (4)$$

Using $p(D_F|D_V)$, the relationship between $L(D_V,D_F)$ and $l(D_V)$ can be written as

$$L(D_V,D_F) = l(D_V)p(D_F|D_V), \quad (5)$$

where $l(D_V)$ is the volume equivalent size distribution of particles.

Inserting Eq. (5) into Eq. (3), we get

$$h(D_F) = \int_{D_{V\text{min}}}^{D_{V\text{max}}(D_F)} l(D_V)p_s(D_{F_s}|D_V)p_o(D_{F_o}|D_V)dD_V. \quad (6)$$

From Eq. (6), we can know that $h(D_F)$ is dependent on intensity distribution and particle shape. Since scattered light fluxes of particles were converted into voltage pulses by an OPC, so that the pulse height distribution $N(v_i)$ is equal to $h(D_F)$. Hence, the pulse height distribution of particles is also dependent on intensity distribution and particle shape.

The pulse height distribution of particles can be measured by an OPC, according to Eq. (2). If we can express the average mass of particles that have the same optical equivalent diameter, then aerosol mass concentration can be attained. In addition, according to Eq. (1), the mass of a spherical particle with signal voltage $v_i$ can be expressed as $k_nv_i^{1.5}$ in the case of intensity uniformity. Under the condition of nonuniform intensity distribution and irregular particles, we introduced the equivalent factor $\alpha$, and then $k_nv_i^{\alpha}$ can be regarded as the average mass of particles with signal voltage $v_i$. In view of volume equivalent diameters of particles that have the same voltage are not equal, if the average density of measured particles is $\rho$, then the average mass of particles with the same voltage in a measurement cycle is given by

$$\overline{M}(v_i) = k\rho v_i^{\alpha} \sum_{j=1}^{\max} L(D_{V_j}, D_F)D_{V_j}^3/N(v_i), \quad (7)$$

where $V_L$ is the sampling rate of the OPC. Hence, the calculation formula for aerosol mass concentration can be expressed as

$$C = k \sum_i N(v_i)v_i^{\alpha}, \quad (8)$$

where $k$ is the proportional coefficient, which has the same dimension with mass concentration.

Equation (8) can be used to calculate aerosol mass concentration only if $N(v_i)$ and $\alpha$ are known. Although $N(v_i)$ could be measured by an OPC, it is difficult to precisely determine $k$ and $\alpha$. Thus, a calibration is performed to determine the unknown coefficients. In previous work\cite{7}, the equivalent factor $\alpha$ was calibrated by the least square method, and $k$ was calculated by

$$k = \frac{1}{T} \sum_{t=1}^{T} \frac{C_{\text{TSL},t}}{\sum_{i=1}^{q} N_t(v_i)v_i^{\alpha}}, \quad (9)$$

where $T$ is the number of experimental data points, $C_{\text{TSL},t}$ is the actual mass concentration at the $t$th measurement.

But detailed analysis of experimental results indicates $\alpha$ is instable, which is related to the range of mass concentration, as shown in Fig. 3. The fluctuation behavior is related to the number of experimental data points. The
less the number of experimental data points, the larger the fluctuations. Hence, the least square method is not fit for calibration of α, however, the calculated results of this method indicates α is basically less than 3. On the other hand, α cannot be less than 0, or else it will lead to the false conclusion that the average mass of particles decreases with the increase of the optical equivalent diameter.

In addition, experimental results show if α is determined, k calculated by Eq. (9) is irrelevant to the range of mass concentration. Hence, in this paper, a novel two-step calibration method is proposed, and the description of the method is summarized as follows. 1) In the case of spherical particles and uniform intensity distribution, α = 1.5. For irregular particles and nonuniform intensity distribution, α must be close to 1.5, so we set α ∈ [0, 3] based on the above analysis. The range of mass concentration Rk is divided into l intervals Rk1, Rk2, ..., Rkl (Rk1 ⊂ Rk2 ⊂ ... ⊂ Rkl). 2) For every α in [0, 3], k is calculated by Eq. (9) for every range of mass concentration, and then using these α and k, mass concentrations are calculated by Eq. (8). Relative errors between calculated and actual mass concentrations are calculated. The ranges αj1, αj2, ..., αjIL that make relative errors within ±10% are found out[8], where j is the times of experiments. 3) Computing the intersection αj = αj1 ∩ αj2 ∩ ... ∩ αjIL, which is the necessary condition that the equivalent factor is irrelevant to the range of mass concentration. In order to make sure the accuracy of the equivalent factor, the experiment was repeated for several times. Then the intersection α = α1 ∩ α2 ∩ ... ∩ αjL is attained. Finding the mean $\bar{\alpha}$ of α, which is the equivalent factor calculated by the two-step calibration method. 4) Inserting $\bar{\alpha}$ into Eq. (9) to calculate k for j times experiments respectively. Finally, computing the mean $\bar{k} = (k_1 + k_2 + ... + k_j)/j$ that is the calibration constant calculated by the two-step calibration method.

According to the above steps, we programmed the method and measured smoke for 5 times using the experimental setup shown in Fig. 4. Measurements were carried out in a super clean room. An aerosol monitor (SIDEPAK AM510, TSI, USA) was used as a norm-referenced instrument. Note that the monitor has been calibrated by the gravimetric method. The principle of this instrument is based on the ensemble-measuring technique[4]. It indicates mass concentration $C_{TSI}$ and stores the data in real time. The OPC displays the pulse height distribution of particles, which is collected by a multi-channel pulse height analyzer (PCI9812, ADlink Tech Inc.). The analyzer has 2048 count channels, and the lower limit of particle size of the OPC is 0.3 µm. Finally, calibration coefficients were worked out by the procedure, and the results are shown in Table 1.

To validate the feasibility of the model, we measured soot and air respectively. Measured pulse height distributions combining calibration coefficients $\bar{\alpha}$ and $\bar{k}$ were applied to calculate mass concentrations, and calculated values were compared with $C_{TSI}$. Figure 5 illustrates the relationship of mass concentrations measured by OPC and the norm-referenced instrument. Linear relationship can be clearly found from the distribution of the data points. Therefore, the experimental data were fitted with linear regression, and fitted equations and coefficient correlations are also shown in Fig. 5. The slopes of the fitted lines are 0.9582 and 0.9220, and the correlation coefficients are 0.9991 and 0.9965, respectively. Obviously, mass concentrations measured by OPC agree well with $C_{TSI}$. Therefore, the model can be applied to calculate aerosol mass concentration with an OPC.

In conclusion, a novel model for inversing aerosol mass concentration with an OPC has been presented. In this
model, the dependences of intensity distribution in the optical sensing volume and particle shape on the pulse height distribution of particles were considered. The model has overcome the instability of the equivalent factor using the two-step calibration method. The experimental results indicate that mass concentrations calculated by the model has high precision. Hence, the model offers a feasible way for an OPC to measure aerosol mass concentration in real time. Note that if there are great differences of properties (e.g. the refractive index, density, component) between measured particles and calibrated particles, the OPC needs to be recalibrated. And if sources of aerosols are various, the model can only give relative mass concentrations, which have a reference value for long-time detection of aerosol mass concentration.

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References