Comment on “Retrieval of Atmospheric Fine Particulate Density Based on Merging Particle Size Distribution Measurements: Multi-instrument Observation and Quality Control at Shouxian” by Li et al.

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Abstract This work raises questions about the omission of Cunningham Slip Correction Factor (\(C_c(D)\)) in geometric equivalent diameter (\(D_p\)) conversions from the electric mobility diameter (\(D_m\)) and the aerodynamic diameter (\(D_a\)) in relation to the calculation of effective density (\(\rho_e\)) as defined by Li et al. (2018; https://doi.org/10.1029/2018JD028956). They reported that \(\rho_e\) could be calculated using the formula “\((D_a/D_m)^2\)”, in which \(C_c(D)\) was not introduced into the conversion between two diameters involving \(D_m\) and \(D_p\) or \(D_a\) and \(D_p\). However, these conversions, which ignored \(C_c(D)\) in the transition regime, are not supported by current studies; nor did the group demonstrate their rationality. This comment shows that an error of up to 24.7% could be caused by ignoring the \(C_c(D)\) during conversions between the \(D_p\) and \(D_m\) for particles with a dynamic shape factor of 2.50 and a \(D_m\) of 550 nm; an error of 5.6% could be caused by ignoring the \(C_c(D)\) during conversions between \(D_p\) and \(D_a\) for particles having a dynamic shape factor of 1.12, a density of 2.20 g/cm\(^3\), and a \(D_a\) of 550 nm. The deviation of \(\rho_e\) could be amplified by 55.5% and 11.2% due to the squared term in the formula. Therefore, this comment argues that it is irrational to ignore \(C_c(D)\) when calculating \(D_p\) and \(\rho_e\) by using \(D_m\) and \(D_a\).

1. Introduction

This is a comment on the recent paper of Li et al. (2018). In that paper, a multi-instrument measurement and quality control scheme was presented to improve the merging algorithm for achieving real-time retrieval of density and to propose a method for estimating the uncertainties of retrieved density. The authors defined the effective density (\(\rho_e\)) as follows:

\[
\rho_e = \frac{\rho}{\chi}
\]  

where \(\chi\) represents the particle dynamic shape factor (DSF) and \(\rho\) represents the particle material density. The \(\rho_e\) could be used for calculating aerosol mass (\(M\)) by multiplying the optical equivalent volume (\(V_{\text{opt}}\)) obtained by remote sensing as follows:

\[
M = V \cdot \rho = V_{\text{opt}} \cdot \rho = V_{\text{opt}} \cdot \rho_e
\]  

where \(V\) represents the particle material volume. The derivation process of \(V\) is shown in equation (3):

\[
V = \frac{\pi}{6} D_p^3 = \frac{\pi}{6} D_m^3 \chi^{-3} \approx \frac{\pi}{6} D_a^3 \chi^{-3} = V_{\text{opt}} \chi^{-3}
\]  

where \(D_p\), \(D_m\) and \(D_a\) represent the geometric equivalent diameter, electric mobility equivalent diameter, and optical equivalent diameter, respectively. Li et al. (2018) also stated that \(D_m\) closely approximated \(D_a\) and gave a conversion between \(D_m\) and \(D_p\) as shown in equation (4):

\[
D_m = D_p \chi
\]  

In addition, \(D_p\) could be converted from aerodynamic diameter (\(D_a\)) using equation (5):

...
where $\rho_0$ represents the unit density of 1.0 g/cm$^3$. They calculated $\rho_e$ in equation (1) by combining equations (4) and (5), which is the primary objective of this comment.

\[
\left( \frac{D_a}{D_m} \right)^2 = \frac{\rho}{\chi^2} = \rho_e
\]

(6)

The calculation of $D_p$ by conversions from $D_m$ and $D_a$ has been applied in many studies. DeCarlo et al. (2004) noted that $D_m$ and $D_a$ could be converted to $D_p$ in the transition regime through equations (7) and (8) as follows:

\[
\frac{D_m}{C_c(D_m)} = \frac{D_p}{C_c(D_p)} \chi
\]

(7)

\[
D_p = D_a \sqrt{\frac{\rho_0 C_c(D_a)}{\rho C_c(D_p)}}
\]

(8)

where $C_c(D)$ is the Cunningham slip correction factor, which must be introduced to account for the reduction in drag that occurs when the relative velocity of the gas at the particle surface is nonzero (Hinds, 1999). The reduced drag is significant when the flow around the particle is in transition and free-molecule regime (Knudsen number, $K_n > 0.1$). $C_c(D)$ was parameterized by Allen and Raabe (1982) and Allen and Raabe (1985) as

\[
C_c(D) = 1 + \frac{\lambda}{D} \left( A + B^* \exp \left( \frac{C^* D}{\lambda} \right) \right)
\]

(9)

where $\lambda$ is the mean free path of the gas molecules, and $A$, $B$, and $C$ are empirically determined constants specific to the system under analysis. $C_c(D)$ has been applied in many studies such as Khlystov et al. (2004), Hand and Kreidenweis (2002), and Hu et al. (2012), and all were cited in the work of Li et al. (2018). However, equations (4) and (5) used in the Li et al. (2018) did not introduce $C_c(D)$ nor did the authors provide any references or arguments for its omission. Here, we compared $D_p$ calculated using equations (4) and (7) with $D_m = 550$ nm and $D_a$ as 550 nm. $D_p$ calculated using equations (5) and (8) with sea-salt particles of $\rho = 2.20$ g/cm$^3$, $\chi = 1.12$, and $D_a = 550$ nm. The former $D_p$ has an error of 24.7%, and the latter has an error of 5.6%. These errors are further amplified when calculating the $\rho_e$ due to the squared term for $D_m$ and $D_a$ in equation (6). These results imply that $C_c(D)$ should not be ignored when converting from $D_m$ and $D_a$ to $D_p$. Given the necessity of introducing $C_c(D)$, the work published by Li et al. (2018) theoretically cannot derive the calculation of the $\rho_e$ shown in equation (6) within an acceptable error range.

2. Methods

Li et al. (2018) showed that the overlap size range for density estimation was generally from a $D_a$ of 400 nm, obtained by aerodynamic particle sizer to a $D_m$ of 594 nm obtained by scanning mobility particle sizer. Thus, it is reasonable for this comment to set $D_m$ and $D_a$ as 550 nm to convert $D_p$. Li et al. (2018) did not measure the DSF of atmospheric particles; they mentioned that ammonium sulfate and soot had $\chi$ values of 1.05 and 2.50, respectively. Therefore, this comment selects three values of 1.10, 1.60, and 2.50 for the conversion from $D_m$ and $D_a$ to $D_p$. Given the necessity of introducing $C_c(D)$, the work published by Li et al. (2018) theoretically cannot derive the calculation of the $\rho_e$ shown in equation (6) within an acceptable error range.

3. Results and Discussion

$K_n$, which determines the flow regime of the gas around a particle, is defined as the ratio of the mean free path ($\lambda$) of gas molecules to the particle radius ($r$) (Baron & Willeke, 2001):
where $D$ represents the particle diameter. When $K_n$ exceeds 0.1 but is less than 10, particles are estimated to be in a transition regime in which flow around the particles significantly reduces the drag force they experience. Therefore, a $C_c(D)$ must be introduced to correct this reduction in the Stokes law (DeCarlo et al., 2004). Li et al. (2018) described that their experiments were carried out in Anhui Province, China (22.7 m above sea level), on 21–26 December 2016. The average temperature was approximately 280 K, and the barometric pressure approximated normal atmospheric pressure; so a calculation for the $\lambda$ of gas molecules yielded 70 nm. Based on that value of $\lambda$ and $D = 550$ nm, the $K_n$ from experiments conducted in the work of Li et al. (2018) was 0.25 and indicated that those experiments were carried out in the transition regime ($K_n > 0.1$). Because of that, $C_c(D)$ should be introduced to conversions from $D_m$ and $D_a$ to $D_p$ theoretically. However, Li et al. (2018) ignored $C_c(D)$ without providing references or rationale for the omission. In the following text, we evaluate the effect of ignoring the $C_c(D)$ in the conversions by theoretical calculations.

Figure 1 shows the differences between the two geometric equivalent diameters by conversions from $D_m$ with $C_c(D)$ ($D_p$) and without $C_c(D)$ ($D_p'$). Specifically, $D_p$ is smaller than $D_p'$ by 9.3, 36.5, and 54.4 nm with DSF values of 1.10, 1.60, and 2.50, respectively. The deviation ratio, defined as the ratio of the difference between $D_p'$ and $D_p$ to $D_p$ ($\frac{(D_p'-D_p)}{D_p}$), was 1.8%, 10.6%, and 24.7% for the different diameters. Li et al. (2018) assumed that there were rare cases of freshly pure elemental carbon (EC) with a DSF as high as 2.5 in the actual atmospheric aerosols. However, Hu et al. (2018) found that fresh EC mixed with fresh organic carbon accounted for 8.2% of atmospheric particles in the Yangtze River Delta by single particle aerosol mass spectrometry, and Li et al. (2017) also observed that freshly pure EC accounted for 21.85% of atmospheric particles in Nanjing, China. These findings imply that it is more reasonable to take the existence of freshly pure EC into consideration in the actual atmosphere. Therefore, $D_p$ would result in an error up to 24.7% in the field observation of Li et al. (2018) when conversing from $D_m$ to $D_p$ without introducing $C_c(D)$.

Table 1 shows the differences between the two geometric equivalent diameters converted from $D_a$ with and without $C_c(D)$. The error ratio was 5.6%. However, Li et al. (2018) calculated $\rho_e$ using equation (6), which included a squared $D_a$ term; this results in amplifying the error into 11.2% for the $\rho_e$. $D_m$ is also squared like $D_a$ in calculations of $\rho_e$; combining the errors from $D_a$ and $D_m$ conversions would cause an unacceptable error of 55.5% for the $\rho_e$ calculation. Therefore, the $C_c(D)$ cannot be ignored in conversions from $D_m$ and $D_a$ to $D_p$.

Based on the introduction of $C_c(D)$, equations (4) and (5) should be replaced by equations (6) and (7). In doing so, equation (3) becomes equation (11):

$$V_n = \frac{\pi}{6} D_n^3 = \frac{\pi}{6} D_m^3 = \frac{\pi}{6} C_c(D_m) \left( \frac{C_c(D_m)}{C_c(D_p)} \right)^3 \frac{C_c(D_m)}{C_c(D_p)} \chi^3 \quad (11)$$

And equation (2) becomes equation (12):

**Table 1**

<p>| Geometric Equivalent Diameter Converted From $D_a$ With $C_c(D)$ and Without $C_c(D)$ |
|---------------------------------|-----------------|-----------------|-----------------|-----------------|</p>
<table>
<thead>
<tr>
<th>$D_a$ (nm)</th>
<th>$D_p$ (with $C_c(D)$; nm)</th>
<th>$D_p'$ (without $C_c(D)$; nm)</th>
<th>Deviation ratio (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>550</td>
<td>371.7</td>
<td>392.4</td>
<td>5.6%</td>
</tr>
</tbody>
</table>
\[ M = V \rho = V n \rho \chi^{-3} \left( \frac{C_c(D_m)}{C_c(D_p)} \right)^{-3} \]  

(12)

Thus, the accurate effective density should be expressed as follows:

\[ \rho_e = \rho \frac{C_c(D_p)}{C_c(D_m)} \]  

(13)

\( C_c(D_p) \) cannot be obtained or replaced by any known variables, indicating that the formula for calculating \( \rho_e \) through \( D_m \) and \( D_a \) presented by Li et al. (2018) cannot derive the \( \rho_e \) with an acceptable error.

4. Conclusions

This comment questions the omission of \( C_c(D) \) in conversions of \( D_p \) from \( D_m \) and \( D_a \) presented by Li et al. (2018). We estimated the \( K_n \) in the work of Z Q Li et al. (2018) and found the sampled aerosols were in the transition regime, which indicated that \( C_c(D) \) should theoretically be introduced to correct the reduction drag force experienced by the particles. The \( D_p \) conversion from \( D_m \) and \( D_a \) with and without \( C_c(D) \) was calculated for the particles with different DSF values and/or density using a diameter (\( D_m \) or \( D_a \)) of 550 nm. Those results showed that neglecting \( C_c(D) \) could cause errors between 5.6 and 24.7% for \( D_p \). In conversions from \( D_m \) and \( D_a \), this error is amplified due to the squared term in the calculations and indicates that the \( C_c(D) \) term in conversions of \( D_p \) from \( D_m \) and \( D_a \) in the transition regime cannot be ignored. Consequently, the method reported in the work of Li et al. (2018) cannot provide \( \rho_e \) values within a reasonable error range.

References


