Supplementary Material for

On the morphology of non-spherical particles using tandem aerodynamic diameter, mobility diameter, and mass measurements

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S1. Uncertainty analysis

The uncertainty analysis of the reported results in the present study was conducted based on ANSI/ASME Measurement Uncertainty Standard (Abernethy et al., 1985). In calculating the total uncertainty, two types of uncertainties should be considered: (i) precision uncertainty (also known as random error) and (ii) bias uncertainty (also known as systematic or instrument error). The precision uncertainty is calculated from repeated measurements and, for a small number of samples (n < 30), is calculated from the following equation

$$P_x = t_{1-c,n-1}\frac{\sigma_n}{\sqrt{n}} \tag{S1}$$

where P_x is the precision uncertainty in quantity x, n is the number of samples (repeated measurements), $t_{1-c,n-1}$ is the Student's t-distribution variable at confidence interval of c (95% in this study) and degree of freedom of $n-1$, and σ_n is the standard deviation of samples (square-root of the variance).

The bias uncertainty, B_x, is the error of the instrument to read the correct value of a measurement. When a reported parameter depends on two or more independent variables as $f(x_1, x_2, x_3, \ldots)$, the propagation of uncertainty is used as follows:

$$B_x = \sqrt{\left(\Delta x_1\frac{\partial f}{\partial x_1}\right)^2 + \left(\Delta x_2\frac{\partial f}{\partial x_2}\right)^2 + \left(\Delta x_3\frac{\partial f}{\partial x_3}\right)^2 + \cdots} \tag{S2}$$

where Δ denotes the uncertainty in the corresponding independent variable. The total uncertainty, U_x, is then estimated as follows

$$U_x = \sqrt{P_x^2 + B_x^2} \tag{S3}$$

The instruments used for the measurement of effective density had the following bias uncertainties: 3% in particle mobility diameter measurement using the DMA (Kinney et al., 1991), 2.7% in mass measurement of singly-charged particles using the CPMA (Symonds et al., 2013), and 4.3% in particle aerodynamic diameter measurement using the AAC (Tavakoli & Olfert, 2014).

To calculate the bias uncertainty in effective density using a combination of any two instruments, the principle of propagation of bias uncertainty was used. Using the DMA-CPMA method, the particle effective density is determined using the following equation as noted in Section 2 of the manuscript

$$\rho_{\mathrm{eff}} = \frac{6m}{\pi d_{\mathrm{m}}^3} \tag{S4}$$

and its bias uncertainty, $\Delta\rho_{\mathrm{eff}}$, is calculated as

$$\frac{\Delta\rho_{\text{eff}}}{\rho_{\text{eff}}} = \sqrt{\left(\frac{\Delta m}{m}\right)^2 + 9\left(\frac{\Delta d_{\text{m}}}{d_{\text{m}}}\right)^2} = \sqrt{(0.027)^2 + 9(0.03)^2} \sim 9.4\% \tag{S5}$$

where Δ denotes the uncertainty (error) in the corresponding physical quantity.

Using the AAC-DMA method, the particle effective density is determined using the following equation as noted in Section 2 of the manuscript

$$\rho_{\text{eff}} = \rho_0 \frac{d_{\text{a}}^2 C_{\text{c}}(d_{\text{a}})}{d_{\text{m}}^2 C_{\text{c}}(d_{\text{m}})} \tag{S6}$$

The mean uncertainty in Cunningham slip correction factor, C_{c}, for particle diameter in the range of 20–270 nm is ~2.5% (Kim et al., 2005). Thus, the bias uncertainty in effective density is calculated as

$$\frac{\Delta\rho_{\text{eff}}}{\rho_{\text{eff}}} = \sqrt{4\left(\frac{\Delta d_{\text{a}}}{d_{\text{a}}}\right)^2 + \left(\frac{\Delta C_{\text{c}}(d_{\text{a}})}{C_{\text{c}}(d_{\text{a}})}\right)^2 + 4\left(\frac{\Delta d_{\text{m}}}{d_{\text{m}}}\right)^2 + \left(\frac{\Delta C_{\text{c}}(d_{\text{m}})}{C_{\text{c}}(d_{\text{m}})}\right)^2} \tag{S7}$$

$$= \sqrt{4(0.043)^2 + (0.025)^2 + 4(0.03)^2 + (0.025)^2} \sim 11.1\%.$$

Using the AAC-CPMA method, the particle effective density is determined using the following equation as noted in Section 2 of the manuscript

$$\rho_{\text{eff}} = \left(\frac{\pi d_{\text{a}}^3}{6m}\right)^2 \left(\frac{\rho_0 C_{\text{c}}(d_{\text{a}})}{C_{\text{c}}(d_{\text{m}})}\right)^3 \tag{S8}$$

and the bias uncertainty in effective density is calculated as

$$\frac{\Delta\rho_{\text{eff}}}{\rho_{\text{eff}}} = \sqrt{36\left(\frac{\Delta d_{\text{a}}}{d_{\text{a}}}\right)^2 + 9\left(\frac{\Delta C_{\text{c}}(d_{\text{a}})}{C_{\text{c}}(d_{\text{a}})}\right)^2 + 4\left(\frac{\Delta m}{m}\right)^2 + 9\left(\frac{\Delta C_{\text{c}}(d_{\text{m}})}{C_{\text{c}}(d_{\text{m}})}\right)^2} \tag{S9}$$

$$= \sqrt{36(0.043)^2 + 9(0.025)^2 + 4(0.027)^2 + 9(0.025)^2} \sim 28.4\%.$$

S2. Technical specifications of the GDI engine

The technical specifications of the GDI engine used in the present study are summarized in Table S1.

Table S1: Engine specifications (bTDC means before Top Dead Center and aTDC means after Top Dead Center)

Specification	Value
Cylinder head	Pentroof type
Compression ratio	12.5:1
Bore	82 mm
Stroke	85 mm
Stroke volume	449 cm3
Fuel direct injection system	Central mounted generic six-hole injector
Injection pressure	150 bars
Spark plug location	Exhaust side
Intake valve timing	Open 334° bTDC Close 166° bTDC
Exhaust valve timing	Open 154° aTDC Close 330° aTDC

S3. Mass-mobility of polydisperse particles

The mass-mobility relationships of non-stripped and stripped polydisperse particles sampled from two engine operating conditions—1200 rpm speed and 12 bar load as well as 2000 rpm speed and 6 bar load—are shown in Figures S1 and S2, respectively. The mass-mobility exponent of non-stripped particles was in the range of ~2.9—~3.0, which implies that these particles were nearly spherical. The mass-mobility exponent of stripped particles was in the range of 2.77–2.80, which indicates that the soot particles without any semi-volatile coating had a compact structure.

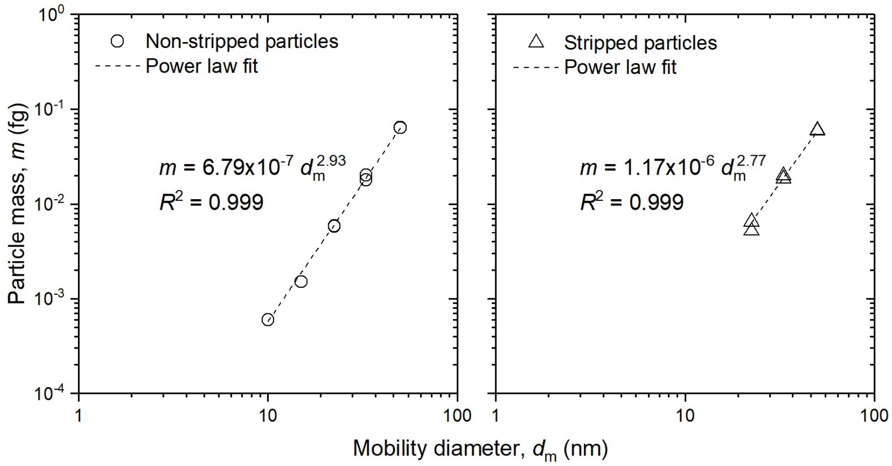

Figure S1: Representative mass-mobility relationship of non-stripped and stripped particles at engine speed of 1200 rpm and load of 12 bar using a DMA to select particles with certain mobility diameters and subsequently measuring their mass using a CPMA.

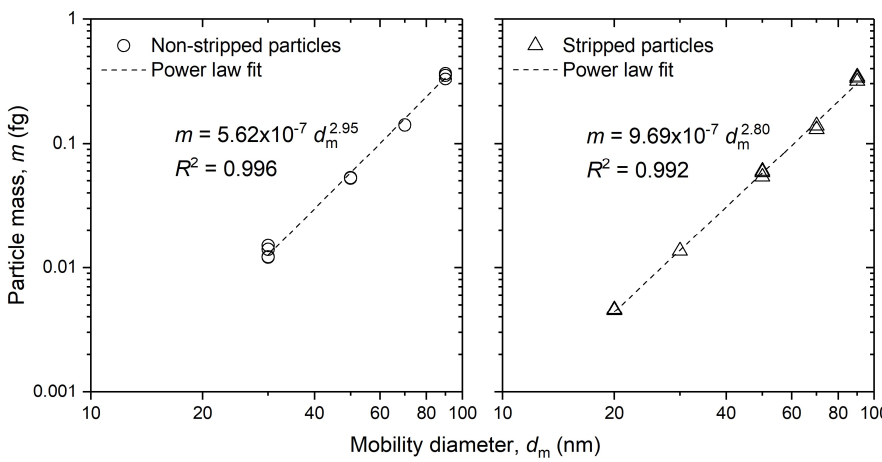

Figure S2: Representative mass-mobility relationship of non-stripped and stripped particles at engine speed of 2000 rpm and load of 6 bar using a DMA to select particles with certain mobility diameters and subsequently measuring their mass using a CPMA.

References

Abernethy, R. B., Benedict, R. P., & Dowdell, R. B. (1985). ASME Measurement Uncertainty. *Journal of Fluids Engineering*, *107*(2), 161–164. https://doi.org/10.1115/1.3242450

Kim, J. H., Mulholland, G. W., Kukuck, S. R., & Pui, D. Y. H. (2005). Slip correction measurements of certified PSL nanoparticles using a nanometer differential mobility analyzer (Nano-DMA) for knudsen number from 0.5 to 83. *Journal of Research of the National Institute of Standards and Technology*, *110*(1), 31–54. https://doi.org/10.6028/jres.110.005

Kinney, P. D., Pui, D. Y. H., Mulholland, G. W., & Bryner, N. P. (1991). Use of the electrostatic classification method to size 0.1 micrometer SRM particles - A feasibility study. *Journal of Research of the National Institute of Standards and Technology*, *96*(2), 147. https://doi.org/10.6028/jres.096.006

Symonds, J. P. R., Reavell, K. S. J., & Olfert, J. S. (2013). The CPMA-electrometer system - A suspended particle mass concentration standard. *Aerosol Science and Technology*, *47*(8). https://doi.org/10.1080/02786826.2013.801547

Tavakoli, F., & Olfert, J. S. (2014). Determination of particle mass, effective density, mass-mobility exponent, and dynamic shape factor using an aerodynamic aerosol classifier and a differential mobility analyzer in tandem. *Journal of Aerosol Science*, *75*, 35–42. https://doi.org/10.1016/j.jaerosci.2014.04.010