The Size, Shape, and Phase of Nanoscale Uric Acid Particles

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Supporting Information

Figure S1: Example droplet size distribution generated after nebulization of aqueous uric acid solution

Figure S2: Example particle size distribution after drying for amorphous uric acid particles
Table S1: Pressure and RH conditions for different drying rates

| ΔP on Shell Side (mm Hg) | RH inlet (%)   | RH exit (%)   |
|--------------------------|----------------|---------------|
| -5                       | 87.9 ± 1       | 22.8 ± 0.2    |
| -10                      | 85 ± 0.6       | 22.4 ± 0.2    |
| -15                      | 89.73 ± 0.6    | 18.9 ± 0.7    |

TEM images and SAED for amorphous uric acid particle

Figure S3: HRTEM and Selected Area Electron Diffraction (SAED) images of droplet dried uric acid particle
Shape factor Calculations:

The shape factor quantifies how close the particle is to a sphere and is derived from the forces acting on the particle. Shape factor value along with volume equivalent diameter is used to correct for non-sphericity of particles. For a particle placed in a fluid flow, forces acting on the particle can be broadly classified into gravitational/body force, the buoyant force due to surrounding media (negligible for a particle in the air) and drag force. We also account for the correction of the drag force due to Brownian motion of surrounding media and the influence of nearby particles. This is done by using a correction term called Cunningham slip correction factor ($C_c$).

The general expression of the drag force acting on a non-spherical particle with a shape factor value $\chi$, volume equivalent diameter $d_{ve}$ and corrected for Brownian motion of surrounding media is given by:

$$F_D = \frac{3\pi\mu v\chi d_{ve}}{C_c(d_{ve})}$$  \hspace{1cm} (S1)

Here $v$ is the velocity of particle relative to surrounding media, and $\mu$ is the viscosity of surrounding media. The Cunningham slip correction factor has the form:

$$C_c(d) = 1 + \frac{2\lambda}{d} \left[ a + b \exp \left( -\frac{c d}{2\lambda} \right) \right]$$  \hspace{1cm} (S2)

Coefficients $a$, $b$ and $c$ have been calculated experimentally by previous researchers (Dahneke 1973a; 1973b) and $d$ is the diameter of particle. The Cunningham slip correction factor is used for small particles where the Brownian motion of surrounding media has a significant impact on the trajectory of the particle under consideration. This correction in drag force is based on the dimension of the particle and the mean free path of the surrounding media. The Knudsen number
\((Kn)\) is referred to as the ratio of the mean free path of surrounding media to the diameter of the particle.

\[
Kn = \frac{2\lambda}{d} \tag{S3}
\]

In the case of mobility diameter calculations, the charged particles are classified by application of the electric field inside a differential mobility analyzer (DMA). Assuming aerodynamic drag and the electrical forces on the particle balance each other out, there will be no acceleration of the particle. Electrical Mobility of an irregular particle with \(n\) charge and with a volume equivalent diameter \((d_{ve})\) is defined as the velocity of migration of particle per unit electrical field.

\[
Z_E = \frac{c_c(d_{ve})n\epsilon}{3\pi\mu \chi d_{ve}} \tag{S4}
\]

Defining mobility diameter of a particle as the diameter of a spherical particle with the same electrical mobility as an irregular particle. Electrical mobility of a particle in terms of mobility diameter can be written as:

\[
Z_E = \frac{c_c(d_{mo})n\epsilon}{3\pi\mu d_{mo}} \tag{S5}
\]

Combining equations S4 and S5, a simplified expression connecting mobility diameter to volume equivalent diameter and shape factor can be written as:

\[
\frac{c_c(d_{ve})}{\chi d_{ve}} = \frac{c_c(d_{mo})}{d_{mo}} \tag{S6}
\]
The aerodynamic diameter of a particle is defined as the diameter of a sphere of density 1000 kg/m³ having the same settling velocity as the particle under consideration. For a non-spherical particle, it is required to take the shape factor of the particle into consideration.

\[
\frac{\pi}{6} \rho_0 d_{ve}^3 g = \frac{3\pi \nu_0 \chi d_{ve}}{C_c(d_{ve})}
\]  

(S7)

Thus, aerodynamic and mobility diameter are connected using the following relationship (Tavakoli and Olfert 2014):

\[
d_{ae} = d_{ve} \sqrt{\frac{C_c(d_{ve}) \rho_p}{\rho_0 C_c(d_{ae}) \chi}}
\]  

(S8)

Equations S6 and S8 are solved simultaneously to obtain volume equivalent diameter and shape factor provided the aerodynamic diameter and mobility diameter are known for the particle. The bulk density of the uric acid (1870 kg/m³) was used to calculate shape factor, which is a source of uncertainty in measured values of shape factor (Tavakoli and Olfert 2013).

**Error Propagation in Shape Factor Calculations:**

Propagation of errors using standard deviations for \( z = x^m y^n \) is given by:

\[
\frac{\Delta z}{z} = \sqrt{\left( m \frac{\Delta x}{x} \right)^2 + \left( n \frac{\Delta y}{y} \right)^2}
\]  

(S9)

The AAC classifies aerosol particles based on their relaxation time, transfer function of AAC can be represented as (Tavakoli and Olfert 2014):
\[
\tau = \frac{2 * Q_{sh}}{4\pi \omega^2 r^2 l}
\]  
(S10)

Using propagation of uncertainty for equation 15:

\[
\left(\frac{\varepsilon_\tau}{\tau}\right)^2 = \left(\frac{\varepsilon_{Q_{sh}}}{Q_{sh}}\right)^2 + 4 \left(\frac{\varepsilon_\omega}{\omega}\right)^2 + 4 \left(\frac{\varepsilon_r}{r}\right)^2 + \left(\frac{\varepsilon_l}{l}\right)^2
\]  
(S11)

Estimated uncertainty for each variable are as following: \(\varepsilon_{Q_{sh}} = 0.1 \frac{L}{min}, \varepsilon_\omega = 5 \text{ rpm}, \varepsilon_l = 2 \text{ mm and } \varepsilon_r = 5 \mu\text{m}\)

This provides us with \(\frac{\varepsilon_\tau}{\tau} = 0.048\)

Relaxation time of a particle is defined as:

\[
\tau = \frac{C_c (d_{ae}) * \rho_0 * d_{ae}^2}{18\pi \mu}
\]  
(S12)

The subsequent error propagation is as follows:

\[
\left(\frac{\varepsilon_{d_{ae}}}{d_{ae}}\right)^2 = 0.25 \left(\frac{\varepsilon_\tau}{\tau}\right)^2 + 0.25 \left(\frac{\varepsilon_\mu}{\mu}\right)^2 + 0.25 \left(\frac{\varepsilon_{C_c}}{C_c}\right)^2
\]  
(S13)

\(\frac{\varepsilon_\mu}{\mu} = 1.2 \%\) assuming temperature uncertainty of 4°C, this gives us

\(\frac{\varepsilon_{d_{ae}}}{d_{ae}} = 2.68 \%\)

\(\tau = Bm\)  
(S14)

The subsequent error propagation is as follows:

\[
\left(\frac{\varepsilon_m}{m}\right)^2 = \left(\frac{\varepsilon_\tau}{\tau}\right)^2 + \left(\frac{\varepsilon_B}{B}\right)^2
\]  
(S15)

For

\[
B = \frac{C_c}{3\pi \mu d_{mo}}
\]  
(S16)

The subsequent error propagation is as follows:
\[ \left( \frac{\varepsilon_B}{B} \right)^2 = \left( \frac{\varepsilon_\mu}{\mu} \right)^2 + \left( \frac{\varepsilon_C}{C_\varepsilon} \right)^2 + \left( \frac{\varepsilon_{dm_0}}{d_{mo}} \right)^2 \]  

(S17)

using \( \frac{\varepsilon_{dm_0}}{d_{mo}} = 3\% \) (Kinney et al., 1991), this gives \( \frac{\varepsilon_B}{B} = 3.8 \% \) and \( \frac{\varepsilon_\mu}{\mu} = 6.1 \% \)

\[ \rho_{eff} = \frac{m}{\left( \frac{\pi}{6} \right) * d_{mo}^3} \]  

(S18)

The subsequent error propagation is as follows:

\[ \left( \frac{\varepsilon_{\rho_{eff}}}{\rho_{eff}} \right)^2 = 9 \left( \frac{\varepsilon_{dm_0}}{d_{mo}} \right)^2 + \left( \frac{\varepsilon_m}{m} \right)^2 \]  

(S19)

\( \frac{\varepsilon_{\rho_{eff}}}{\rho_{eff}} \) was found to be 10.87 %

\[ \chi = \left( \frac{d_{mo}}{d_{ae} * C_C(d_{mo})} \right)^{2/3} \frac{1}{\rho_{p}^5 C_C(d_{ae})} \]  

\( \frac{1}{\rho_{p}^5} \)

(S20)

The subsequent error propagation is as follows:

\[ \left( \frac{\varepsilon_\chi}{\chi} \right)^2 = \frac{4}{9} \left( \frac{\varepsilon_{dm_0}}{d_{mo}} \right)^2 + \frac{4}{9} \left( \frac{\varepsilon_{dae}}{dae} \right)^2 + \frac{14}{9} \left( \frac{\varepsilon_C}{C_C} \right)^2 + \frac{1}{9} \left( \frac{\varepsilon_{\rho_p}}{\rho_p} \right)^2 \]  

(S21)

\( \varepsilon_{\rho_p} \) values for DOS and soot are \( 0.003 \, \frac{g}{cm^3} \) and \( 0.07 \, \frac{g}{cm^3} \) respectively (Park et al., 2004a, 2004b),

we assume uric acid will have a value intermediate of 0.03 and assuming \( \frac{\varepsilon_C(d_{ae})}{C_C(d_{ae})} = \frac{\varepsilon_C(d_{mo})}{C_C(d_{mo})} = \frac{\varepsilon_C(d_{ve})}{C_C(d_{ve})} = 2.1 \% \) (Allen and Raabe, 1985). The final values of \( \frac{\varepsilon_\chi}{\chi} \) was found to be 5.21 %