Application of the stochastic particle-resolved aerosol model PartMC to chamber experiments

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Introduction

The stochastic particle-resolved aerosol model PartMC is a recently developed aerosol model that explicitly resolves and tracks the size and composition of individual particles as they undergo transformations by coagulation and condensation in the atmosphere (Riemer et al., 2009). For this study we adapted PartMC to represent the aerosol evolution in aerosol chamber environment by including chamber-specific processes and treatment of fractal agglomerates based on Naumann (2003). Then code was then validated by comparing PartMC predictions against experimental data obtained from an aerosol chamber at the Department of Civil and Environmental Engineering at the University of Illinois.

Model set-up

Governing equation. The differential equation governing the time evolution of the aerosol size distribution in the chamber can be written as Eq. 1 (Naumann, 2003).

$$\begin{aligned}\frac{\partial n(R_{\mathrm{m},k},t)}{\partial t} = &\underbrace{S_k(t)}_{\text{source rate}} - n(R_{\mathrm{m},k},t)[\underbrace{\alpha_k^{\mathrm{D}}(t)}_{\text{wall loss}} + \underbrace{\alpha_k^{\mathrm{S}}(t)}_{\text{sedimentation}} + \underbrace{\alpha_k^{\mathrm{L}}(t)}_{\text{leakage}}] \\ &\underbrace{- n(R_{\mathrm{m},k},t)\sum_{i=1}^{N_\mathrm{s}}\left(1-\frac{\delta_{i,k}}{2}\right)K_{ik}n(R_{\mathrm{m},i},t)}_{\text{coagulation loss}} \\ &\underbrace{+\sum_{i=1}^{N_\mathrm{s}}\sum_{j=i}^{N_\mathrm{s}}K_{ij}n(R_{\mathrm{m},i},t)n(R_{\mathrm{m},j},t)\beta_{ij}^{k}\delta_{k,i+j}}_{\text{coagulation gain}}\end{aligned} \quad (1)$$

The Brownian coagulation kernel K_{ij} has been adjusted to account for fractal particle dynamics based on fractal dimension d_{f}.

Wall diffusion and sedimentation loss in chamber. Define A_{D} as diffusional deposition area, A_{S} as sedimentational deposition area, R_{me} as the particle mobility equivalent radius, R_{m} as mass equivalent radius, the deposition coefficient for wall diffusion is given by

$$\alpha_i^D = \frac{D(R_{\mathrm{me},i})A_{\mathrm{D}}}{\delta_{\mathrm{D}}V} \quad (2)$$

where δ_{D} is the diffusive boundary layer thickness.

$$\delta_{\mathrm{D}} = k_{\mathrm{D}}\left(\frac{D}{D_0}\right)^{\mathrm{a}} \quad (3)$$

The deposition coefficient for sedimentation is

$$\alpha_i^S = \frac{4\pi\rho R_{\mathrm{m},i}^3 g D(R_{\mathrm{me},i})A_{\mathrm{S}}}{3kTV} \quad (4)$$

Using these expressions, PartMC simulates the evolution of the particle dynamics by stochastic sampling.

Fitting optimization approach. To constrain the unknown parameters k_{D}, a and d_{f} in the governing equation, we developed a fitting optimization approach to determine the best-estimate values based on minimizing the L2-norm of the model errors of number and mass distributions.

Model validation

Experimental set-up. The measurements were conducted under ambient conditions in Department of Civil and Environmental Engineering in University of Illinois ($\mathrm{V} = 0.21\ \mathrm{m}^3$, $\mathrm{A_D} = 2\ \mathrm{m}^2$, $\mathrm{A_S} = 0.25\ \mathrm{m}^2$), as shown in Figure 1.

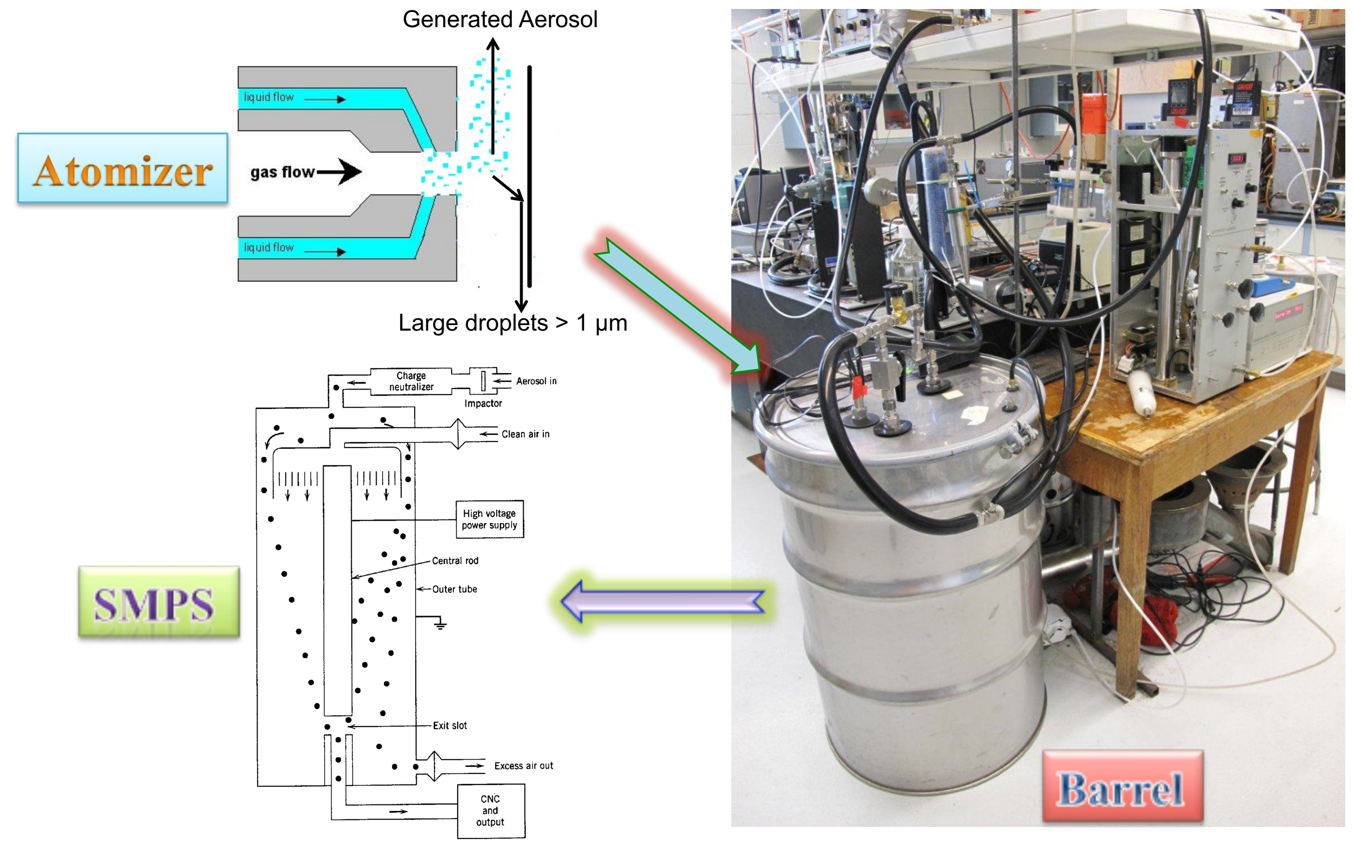

Figure 1: Schematic of experimental set-up of UIUC chamber.

Spherical dry ammonium sulfate particles were introduced to the chamber and experienced coagulation, dilution and wall loss during the evolution.

Scanning electron microscope (SEM) images of particle filter. Figure 2 shows the SEM images of the filters at the end of the measurement, which suggest that coagulation significantly altered the particle structure from primary spherical particles to fractal-like agglomerates.

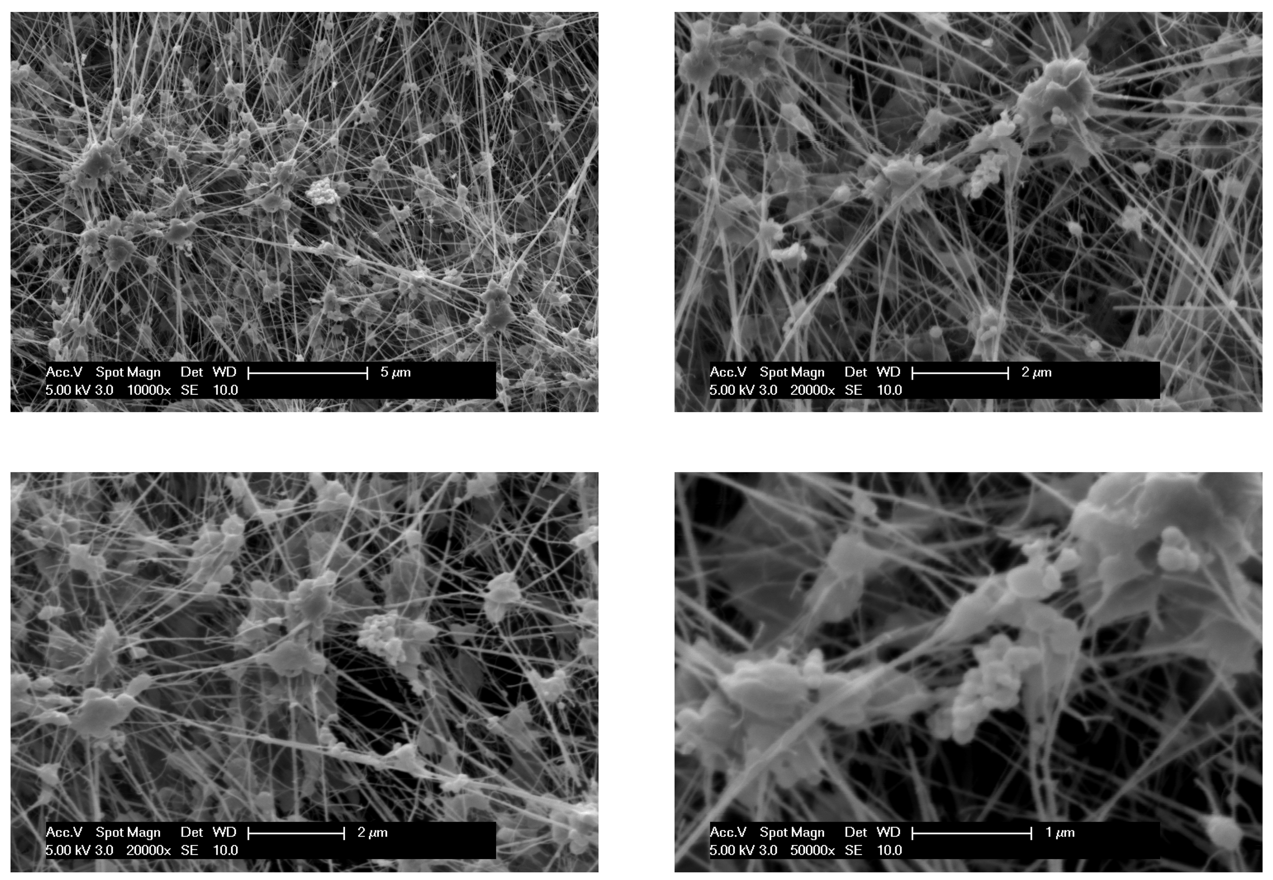

Figure 2: Scanning electron microscope (SEM) images of particle filter at different resolutions. The stripes represent the fibers of the filter.

Comparison of size distributions between model simulation and measurement. The best-estimate values of wall loss parameters and fractal dimension ($k_{\mathrm{D}} = 0.03$, $a = 0.23$, $d_{\mathrm{f}} = 2.6$) were determined by combining three different chamber experiments with varying experimental conditions. This best-estimate was then applied to the 4th experimental dataset to validate the implementation of wall loss and fractal particle treatment in PartMC model.

Figure 3 shows the comparison of number distributions when we use spherical particle treatment ($d_{\mathrm{f}} = 3$, left panel) as well as fractal particle treatment with best-estimate values ($d_{\mathrm{f}} = 2.6$, right panel). It suggests that non-spherical structure of the particles should be considered in model simulation in order to obtain good agreement with measurement results.

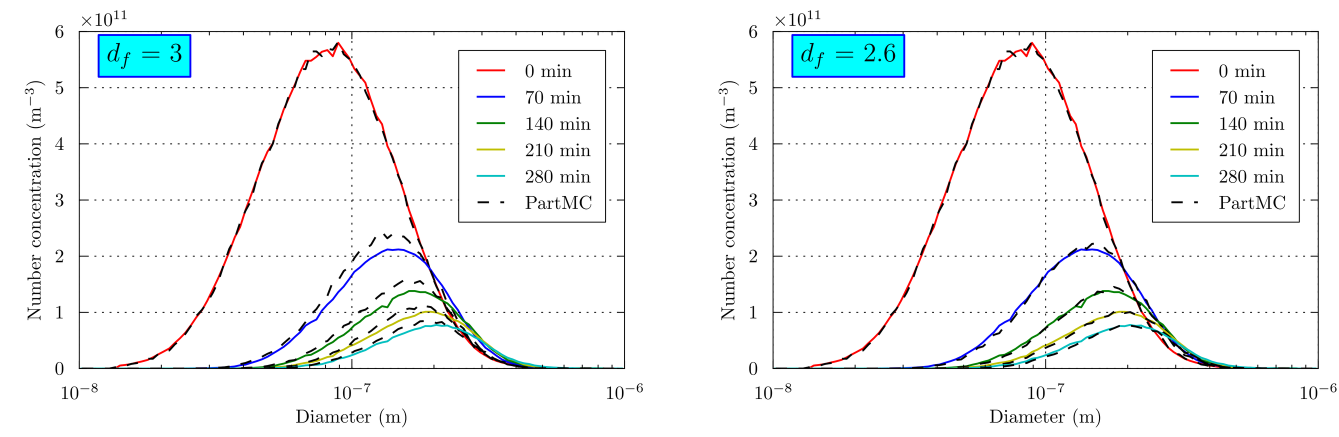

Figure 3: Number distribution comparison between simulation and measurement for (left), spherical particle treatment, and (right), fractal particle treatment in the model.

Similar results can also be observed from Figure 4, which shows the comparison of mass distributions.

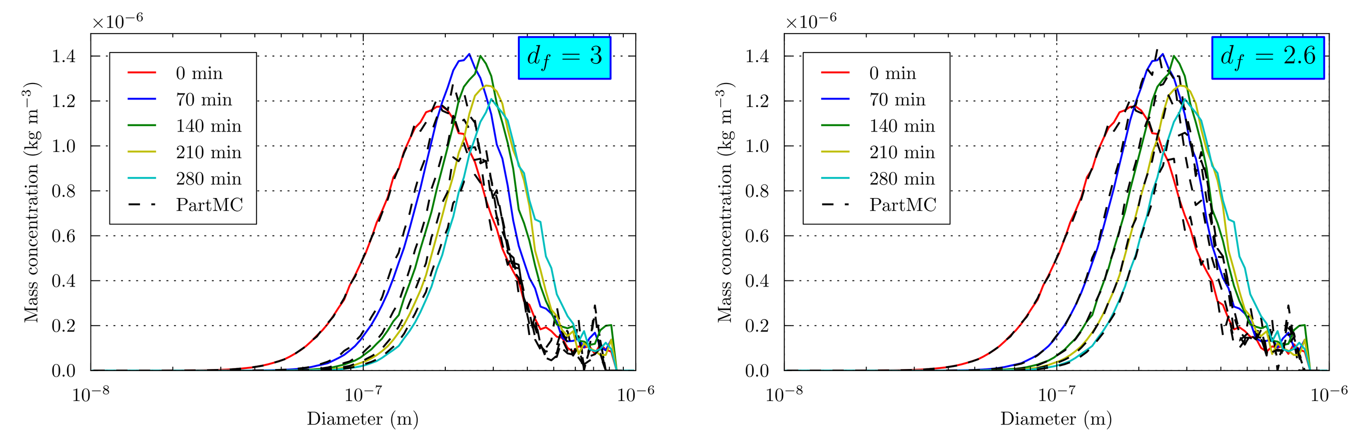

Figure 4: Same as Figure 3, but for mass distributions.

Conclusions

- We implemented a treatment of fractal aerosols and wall loss in PartMC to simulate the evolution of the dynamics of particles in the UIUC chamber.
- The SEM images and model-measurement comparisons emphasized that we need to take into account the non-spherical particle structure during their evolution in the chamber.
- The model framework based on this study will help to interpret and guide chamber experiments in the future.

References

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