A new moment-based parameterization for organic aerosols (MOMSOA): application to a case study at Mt Tai in China

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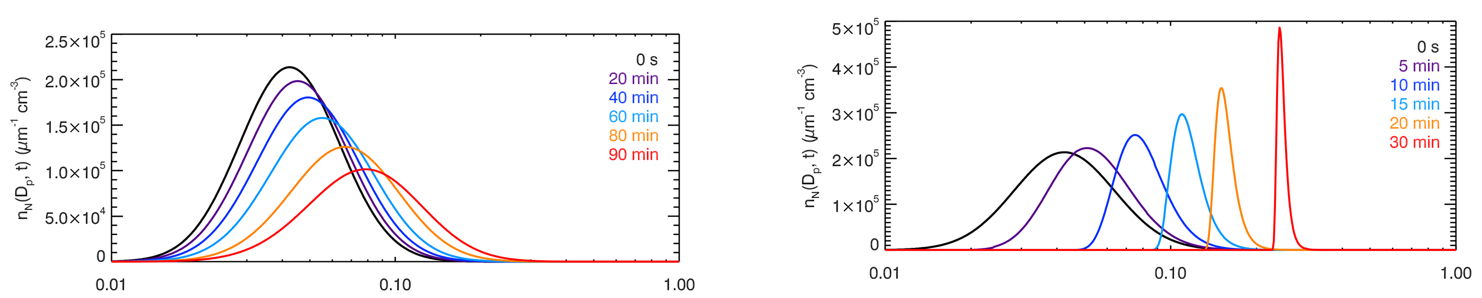
Abstract Secondary organic aerosols (SOA) account for a significant fraction of submicron aerosols. SOA formation changes the preexisting aerosol size distribution, which affects the aerosol direct and indirect radiative forcing, but currently there are no tools to simulate this evolution of aerosol size distribution. Here we developed a new moment-based parameterization scheme, MOMSOA, to describe the evolution of aerosol size distribution due to SOA formation. The scheme is highly computationally-efficient without losing detailed physicochemical information. We implemented MOMSOA into WRF-Chem model to improve the model ability to simulate SOA. From June 13th to June 21th, the model simulates aging of SOA precursors and resulting new particle formation and evolution of aerosol size distribution. Our new parameterization scheme predicts higher OA concentrations than the RACM-SOA-VBS scheme in WRF-Chem, and agrees better with measurements of size distribution evolution due to aerosol aging compared to the default RACM-SOA-VBS scheme.

1. Motivation: SOA formation mechanisms govern aerosol size distribution evolution

Quasi-equilibrium growth
Volume-determined, Raoult's law

Diffusion-limited growth
Surface-determined, no Raoult's law

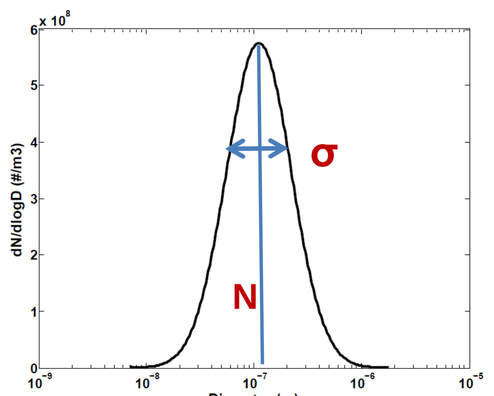


Source:
Zhang et al. 2012

- New laboratory results showed that SOA is formed kinetically by **diffusion-limited growth** (Riipinen et al., 2011; Perraud et al., 2012)
- Current chemical models usually describe SOA formation using absorptive partitioning theory, assuming instantaneous equilibrium between gas and particle phases and only consider the SOA mass.

Our idea: based on the diffusion-limited growth process, we developed a highly computationally-efficient moment-based parameterization scheme to describe aerosol size evolution.

2. Moment-based parameterization scheme development



D_m

$M_p = \int D^p \cdot n(D)dD$

M_0 = const (N)

$M_2 = \int D^2 \cdot n(D)dD$

$M_3 = \int D^3 \cdot n(D)dD$

D_m, σ, N ⟷ M_0, M_2, M_3

- M_0, M_2, M_3 could represent the aerosol size distribution as D_m, σ, N equally, and are directly linked to aerosol climate effects
- We parameterize the time tendency of the three moments to track aerosol size distribution evolution.

I. Use 200 bins to describe aerosol size distribution. Integrate the time tendency of 2nd and 3rd moment.

The increase rate of 2nd and 3rd moment is determined by the diffusion-limited growth.

K: aerosol bins S: organic vapors with different volatility

$$\frac{dM_2}{dt}=\frac{8D_iM_i}{RT\rho}\sum_{k,s}\Bigg(P_s*N_k*\underbrace{\frac{1}{1+\frac{2\lambda}{\alpha D_k}}}_{f1}-P_{eqs}\underbrace{\frac{As(D_k)}{\sum_s As(D_k)}\exp\left(\frac{4\sigma M_i}{RT\rho D_k}\right)\frac{1}{1+\frac{2\lambda}{\alpha D_k}}}_{f2}\Bigg)$$

$$\frac{dM_3}{dt}=\frac{12D_iM_i}{RT\rho}\sum_{k,s}\Bigg(P_sN_kD_k\underbrace{\frac{1}{1+\frac{2\lambda}{\alpha D_k}}}_{f3}-P_{eqs}N_kD_k\underbrace{\frac{As(D_k)}{\sum_s As(D_k)}\exp\left(\frac{4\sigma M_i}{RT\rho D_k}\right)\frac{1}{1+\frac{2\lambda}{\alpha D_k}}}_{f4}\Bigg)$$

f1, f2, f3, f4 are all size-related factors.

II. Parameterize f1, f2, f3, f4 numerically

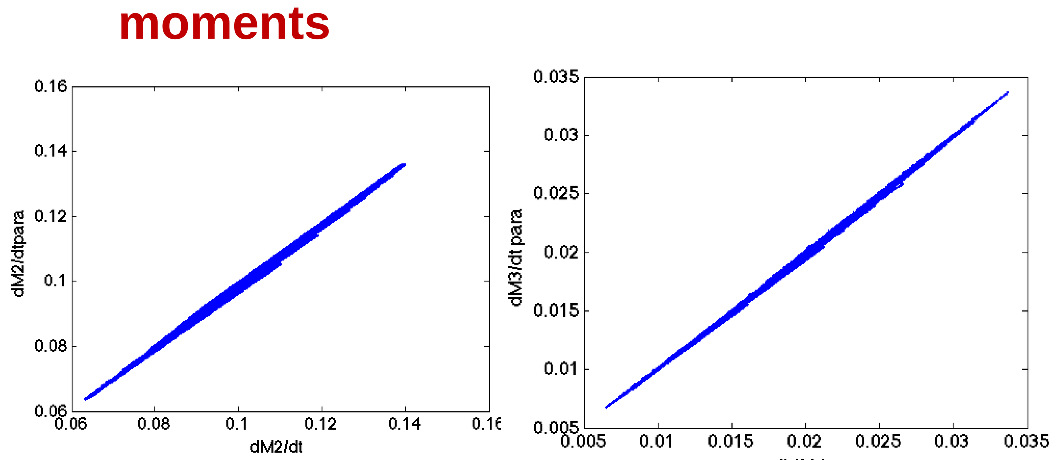
We first use detailed aerosol bins to calculate the four factors and then parameterize them as the function of M_0, M_2, M_3. Take f1 as an example:

$$f1=\sum_k N_k*\frac{1}{1+\frac{2\lambda}{\alpha D_k}}==\exp(-0.21-0.11*\log(D_m)^2-0.03*\log(\sigma)^2)$$

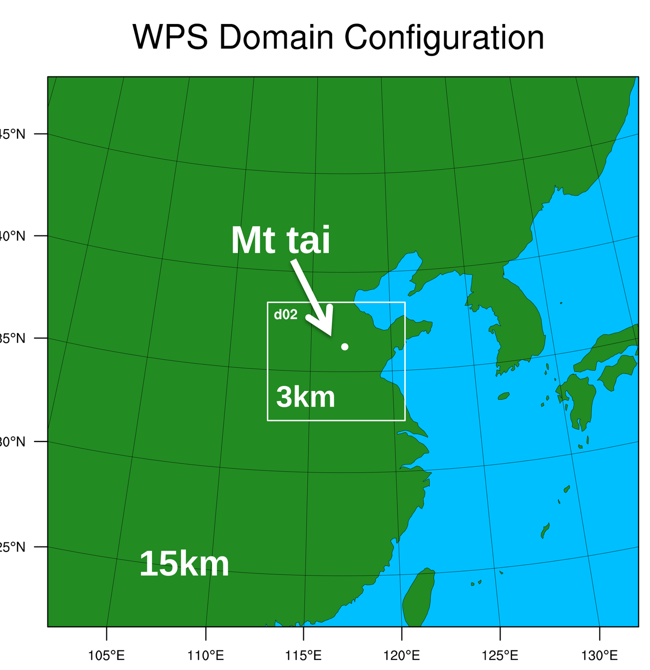
For log-normal size distribution, D_m, σ is calculated by M_0, M_2, M_3

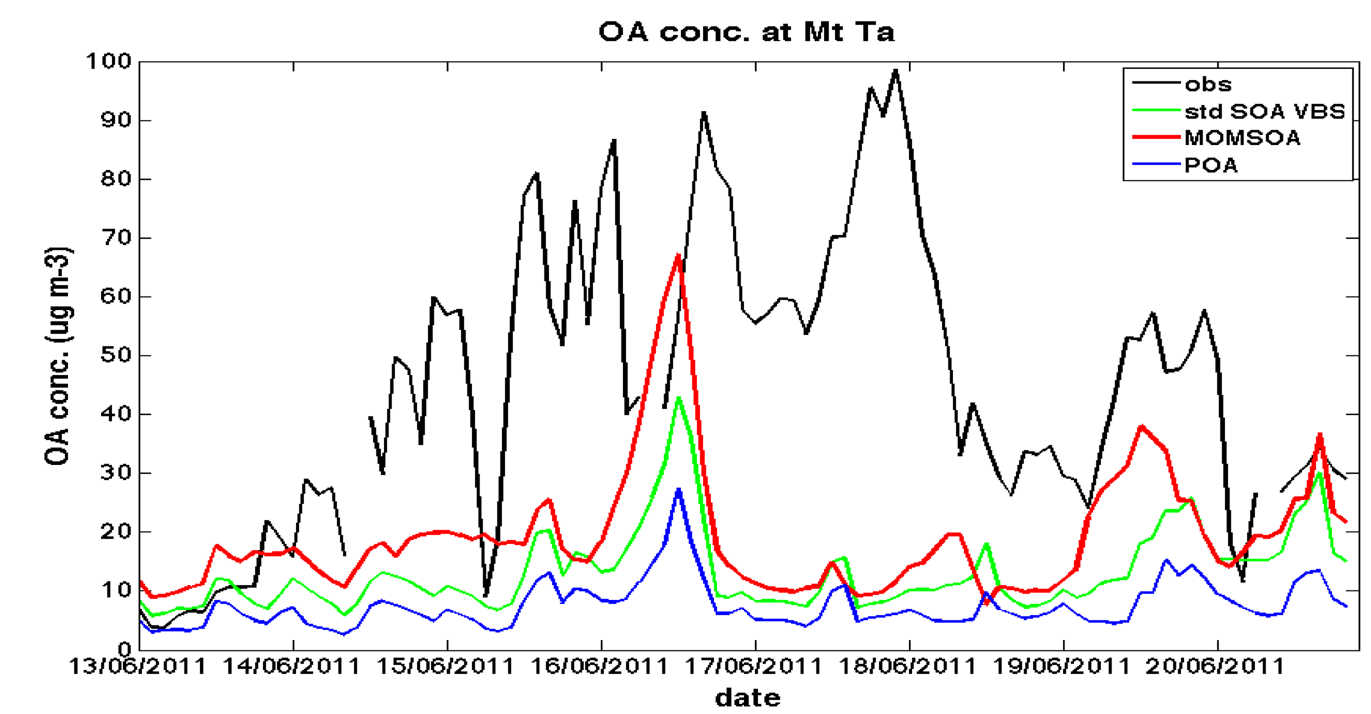
$$D_m=\sqrt[3]{\frac{M_3}{M_0\exp(4.5\sigma^2)}} \qquad \sigma=\sqrt{\frac{1}{3}(\log(M_0)+2\log(M_3))-\log(M_2)}$$

III. Comparison of parameterized and bin-calculated time tendency of 2nd and 3rd moments



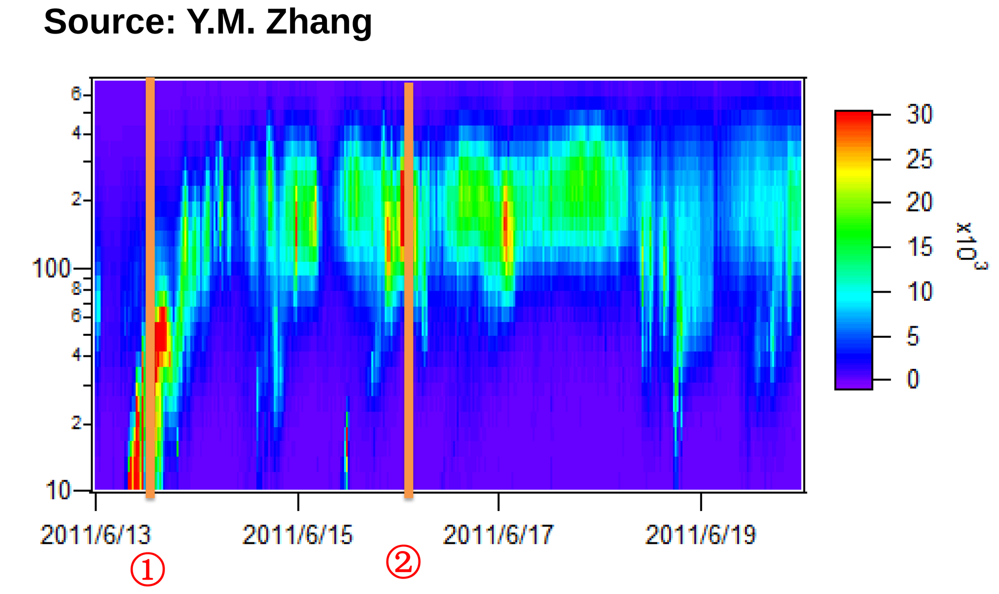
3. Mt Tai case study: comparison w/ observation & the RACM-SOA-VBS scheme in WRF-Chem



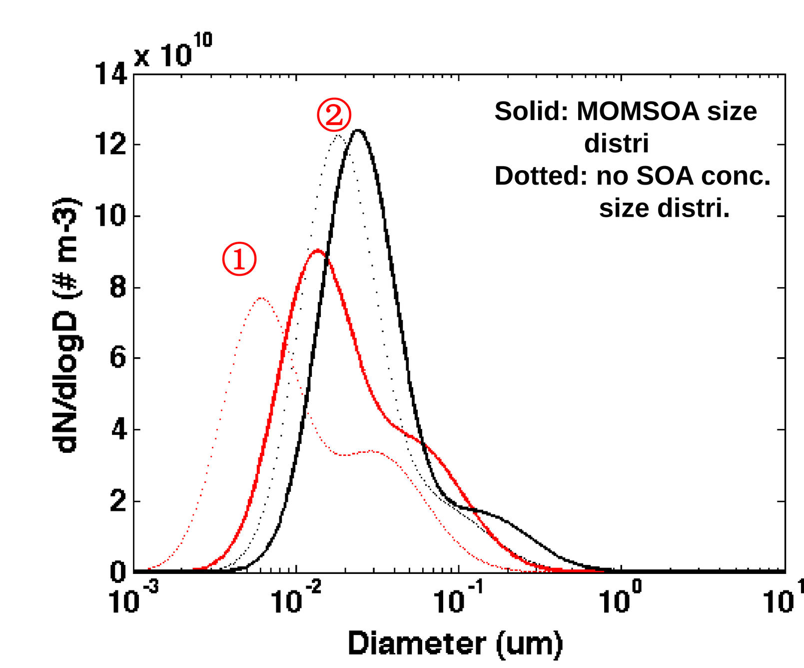


- Simulation: WRF-Chem simulation for organic aerosols at Mt Tai during June 8-21, 2011
- Observation: Size-resolved organic aerosol conc. by AMS and aerosol number conc. by SMPS during June 13-21, 2011
- Compared with WRF-Chem's standard SOA VBS scheme: MOMSOA predicted higher SOA concentrations, closer to the observations
- The computation cost of MOMSOA is comparable with std SOA VBS in WRF-Chem, but with better model performance
- The large discrepancy between observed and simulated OA is mainly due to poor spatial/temporal representation of OA emissions from biomass burning

Observed aerosol number size distribution at Mt Tai
Source: Y.M. Zhang



Modeled aerosol number size distribution at Mt Tai



- Our parameterization reasonably captured the size distribution difference for aerosol aging process. From June 13th to June 16th, the median diameter grows from **0.015 μm to 0.025 μm**
- The difference between solid and dotted lines shows SOA contribution to aerosol size distribution. SOA formation makes aerosol size grow larger.
- ① median diameter: **0.007 to 0.015** μm
- ② median diameter: **0.02 to 0.025** μm

Take home message: We developed a moment-based computationally efficient parameterization to improve simulated impacts of SOA on the evolution of aerosol size distribution

References:

1. Chen et al. (2013): A statistical–numerical aerosol parameterization scheme. ACP
2. Zhang et al. (2014): Chemical composition and mass size distribution of PM1 at an elevated site in central east China. ACP