TAMPERE UNIVERSITY OF TECHNOLOGY

Sulfur Driven Nucleation in Diesel Exhaust: Simulations of a Laboratory Sampling System

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Background

Sulfur driven nucleation in diesel exhaust

- **The amount of sulfuric acid is connected to volatile nucleation mode particle concentration**
  - Modeling studies lack of quantitative information on nucleation rate
  - Dependence of nucleation rate J on gaseous sulfuric acid concentration $[H_2SO_4]$:

$$J \propto [H_2SO_4]^n$$

  where n, i.e. the nucleation slope varies depending on the theory
  - Classical nucleation theory is currently the only one that provides the nucleation rate quantitatively
    - Parameterization of homogeneous binary water-sulfuric acid nucleation rate by Vehkamäki et al. (2002, 2003)
    - The nucleation slope is 5 or more

Exhaust sampling system

- **Partial flow sampling with porous tube type primary diluter and ageing chamber**
- **Observed to mimic real-world nucleation of diesel exhaust**
- **Measured by Rönkkö et al. (2013):**
  - Gaseous sulfuric acid concentrations in raw exhaust
  - Particle distributions after the ageing chamber

Model

Fluid dynamics

- **Steady state finite volume method**
  - ANSYS FLUENT 14.0 CFD-solver
  - Fluid flow, turbulence, heat, and gas transport modeling

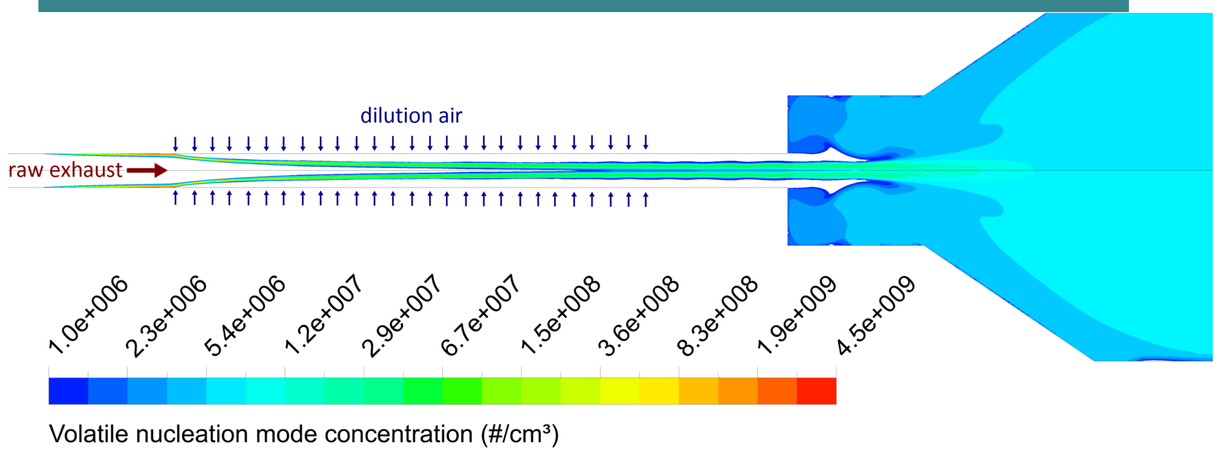
Aerosol dynamics

- **Modal aerosol dynamics code**
  - Coupled with fluid dynamics modeling
- **Transport equation of a moment M_k**

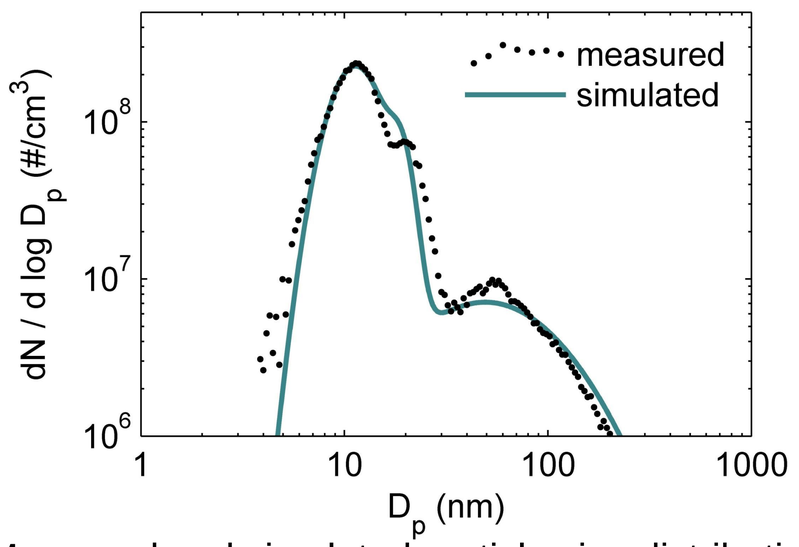
$$\frac{\partial M_k}{\partial t} = -\nabla \cdot (M_k \boldsymbol{u}) + \nabla \cdot \left(\rho_f \overline{D}_{eff,k} \nabla \frac{M_k}{\rho_f} \right) + nucl_k + cond_k + coag_k$$

- **Nucleation**
  - Classical homogeneous binary H_2SO_4-H_2O nucleation rate multiplied by a correction factor
- **Condensation**
  - Sulfuric acid, water, and a wide scale of hydrocarbons are considered as the condensing species
- **Coagulation**
  - Coagulation between different modes

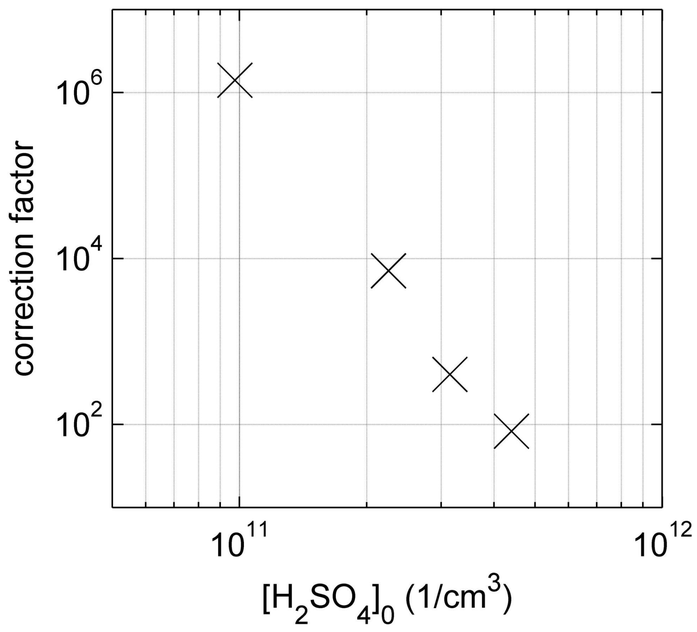
Simulation results



- **Fitting the simulated particle distributions with the measured ones**
  - Adjusting the correction factor → number concentration
  - Adjusting the hydrocarbon amount in raw exhaust → particle size



Measured and simulated particle size distributions



Correction factor versus sulfuric acid concentration in raw exhaust

Conclusions

Correction factor

- **High values**

→ The theory predicts too low concentrations

- **Large variation**

→ May indicate that the use of classical nucleation theory is impractical to model the particle formation in vehicle exhaust

- **Decreasing exponentially**

→ The nucleation slope may be overestimated

→ Other compounds may participate in nucleation

Acknowledgements

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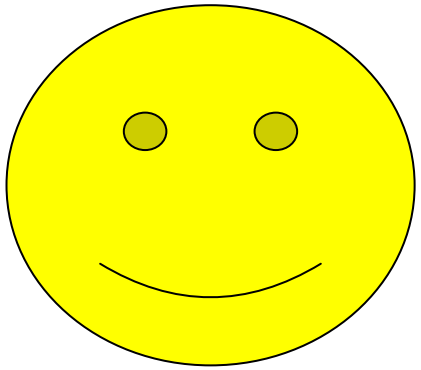

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