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 $[\text{H}_2\text{SO}_4]$:
  $$ J \propto [\text{H}_2\text{SO}_4]^n $$
  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 $M_k$
  $$ \frac{\partial M_k}{\partial t} = -\nabla \cdot (M_k \mathbf{u}) + \nabla \cdot \left( \rho_{ij} D_{ij} \nu M_k \frac{M_k}{\rho_{ij}} \right) + \text{nucl}_k + \text{cond}_k + \text{coag}_k $$
  - Nucleation
    - Classical homogeneous binary $\text{H}_2\text{SO}_4$-$\text{H}_2\text{O}$ 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 $\rightarrow$ number concentration
  - Adjusting the hydrocarbon amount in raw exhaust $\rightarrow$ particle size

Conclusions

Correction factor

- High values
  $\rightarrow$ The theory predicts too low concentrations
- Large variation
  $\rightarrow$ May indicate that the use of classical nucleation theory is impractical to model the particle formation in vehicle exhaust
- Decreasing exponentially
  $\rightarrow$ The nucleation slope may be overestimated
  $\rightarrow$ Other compounds may participate in nucleation

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