



## 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  $M_k$**

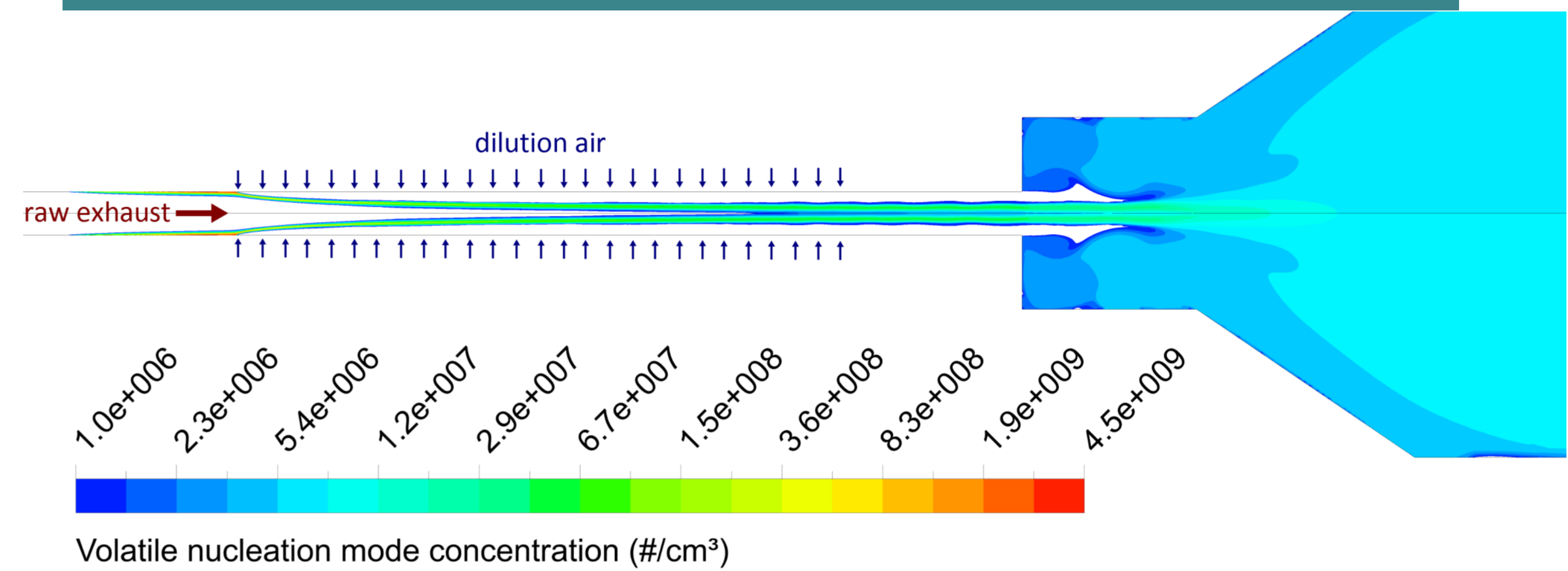
$$\frac{\partial M_k}{\partial t} = -\nabla \cdot (M_k \mathbf{u}) + \nabla \cdot \left( \rho_f \bar{D}_{eff,k} \nabla \frac{M_k}{\rho_f} \right) + nucl_k + cond_k + coag_k$$

- **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

### Acknowledgements

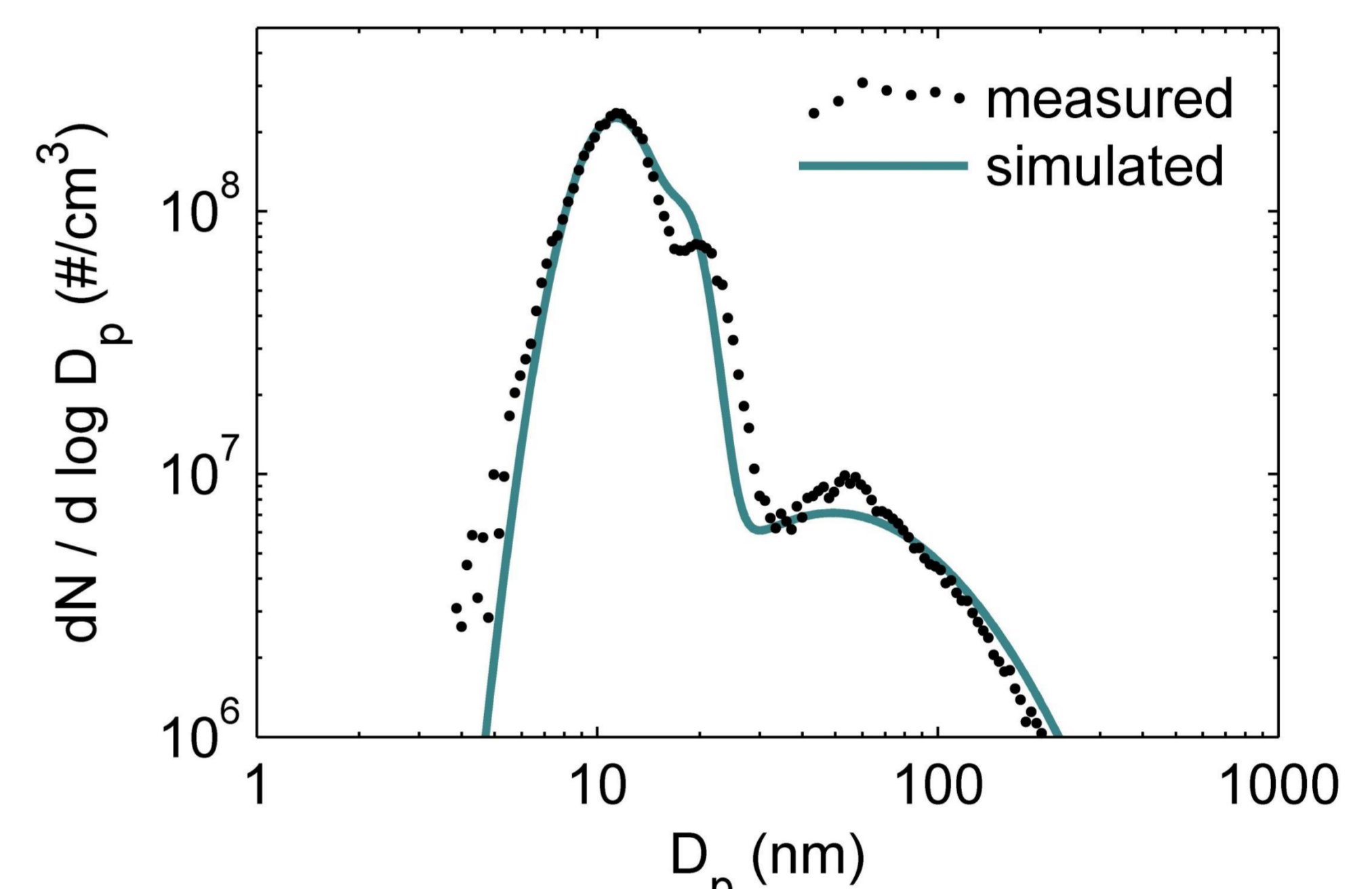
The work is funded by Maj and Tor Nessling Foundation

## Simulation results

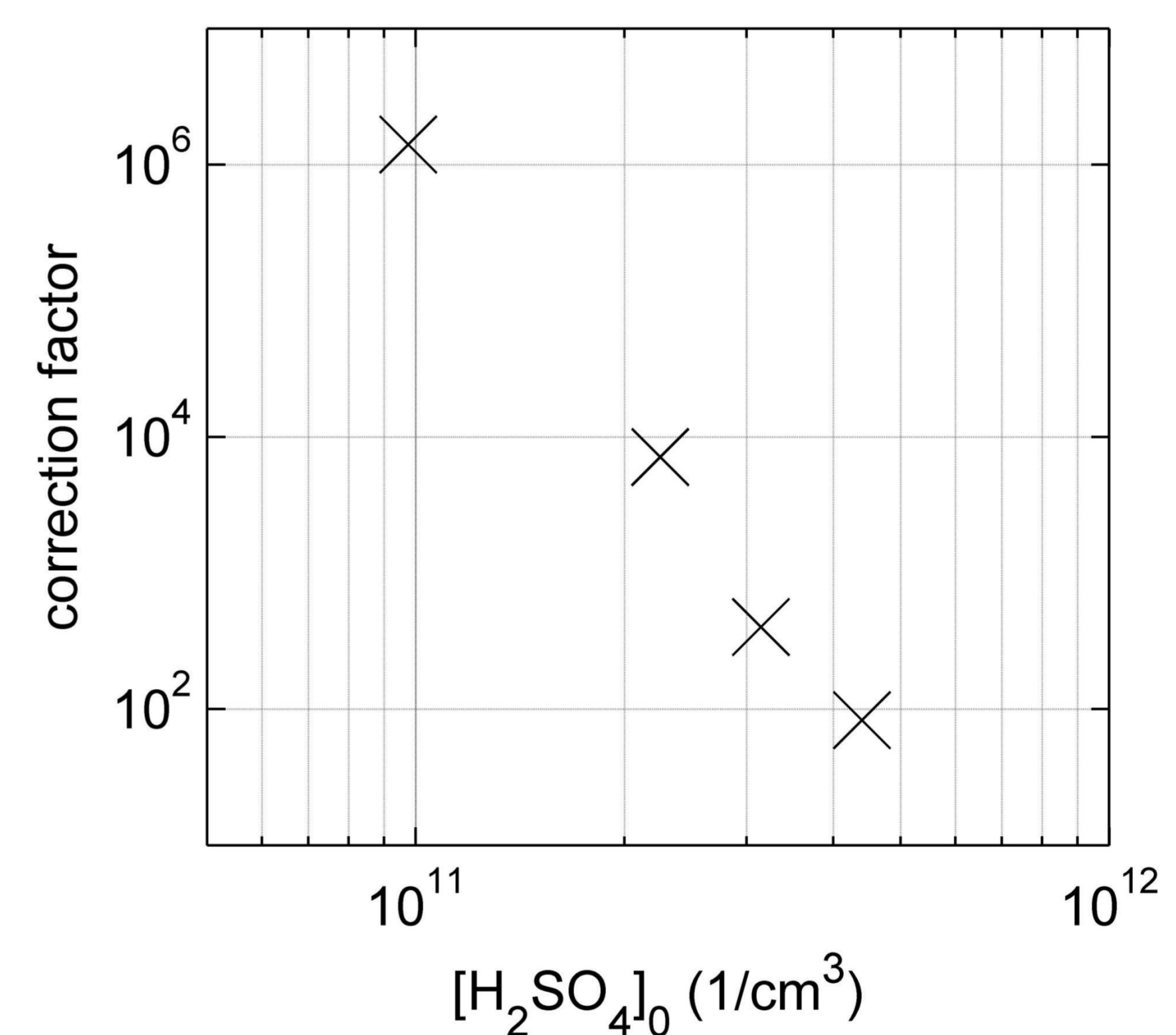


- **Fitting the simulated particle distributions with the measured ones**

- 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

### References

- T. Rönkkö *et al.*, Environ. Sci. Technol. **47**, 11882 (2013)
- H. Vehkamäki *et al.*, J. Geophys. Res. **107**, 4622 (2002)
- H. Vehkamäki *et al.*, Environ. Sci. Technol. **37**, 3392 (2003)