# Sensitivity of Concentration of Accumulation-mode Aerosol Particles to the Representation of New Particle Formation and Particle Emissions in Chemical Transport Models

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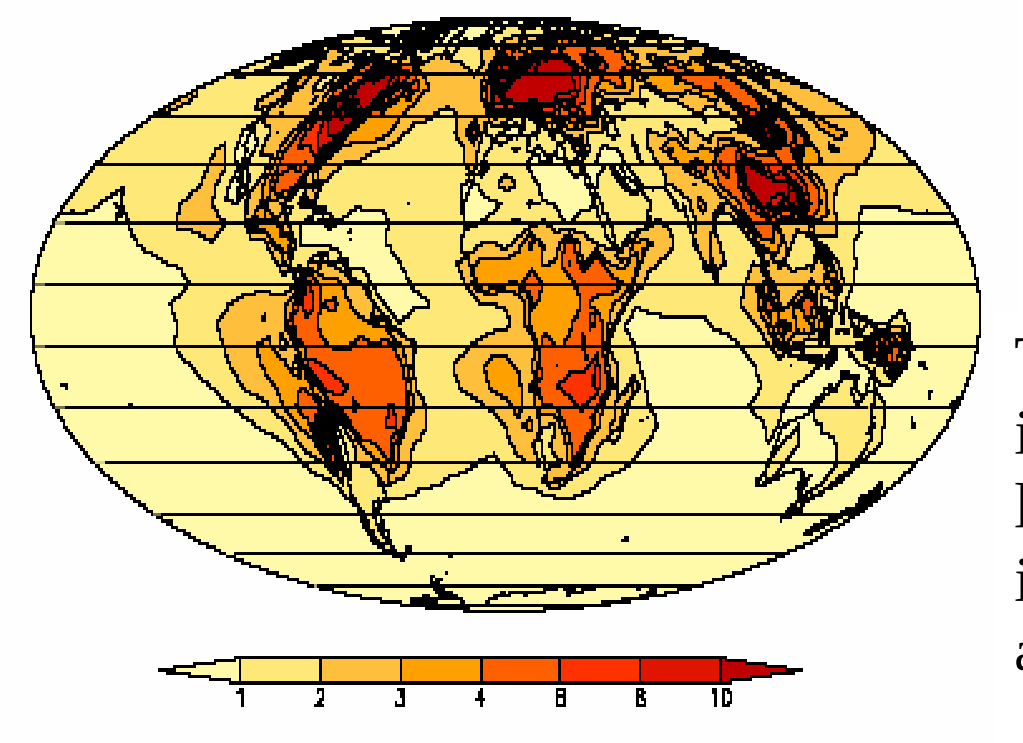
# UNCERTAINTY OF AEROSOL INDIRECT FORCING




Uncertainty of aerosol indirect forcing (W $m^{-2}$)

The uncertainty of aerosol indirect forcing is very large, in large part because of uncertainty in number concentration ( Y.Chen and J.E. Penner, *A.C.P.*, 2005 )

# KEY SENSITIVITIES EXAMINED

- Contribution of nucleation by the ion-ion recombination (small but widespread)
- Enhancement to nucleation rate due to $NH_3$
- Parameterizations for the binary nucleation rate
- Mean diameter of primary particles
- Scheme to grow Aitken mode particles into the accumulation mode

# MODEL VARIANTS

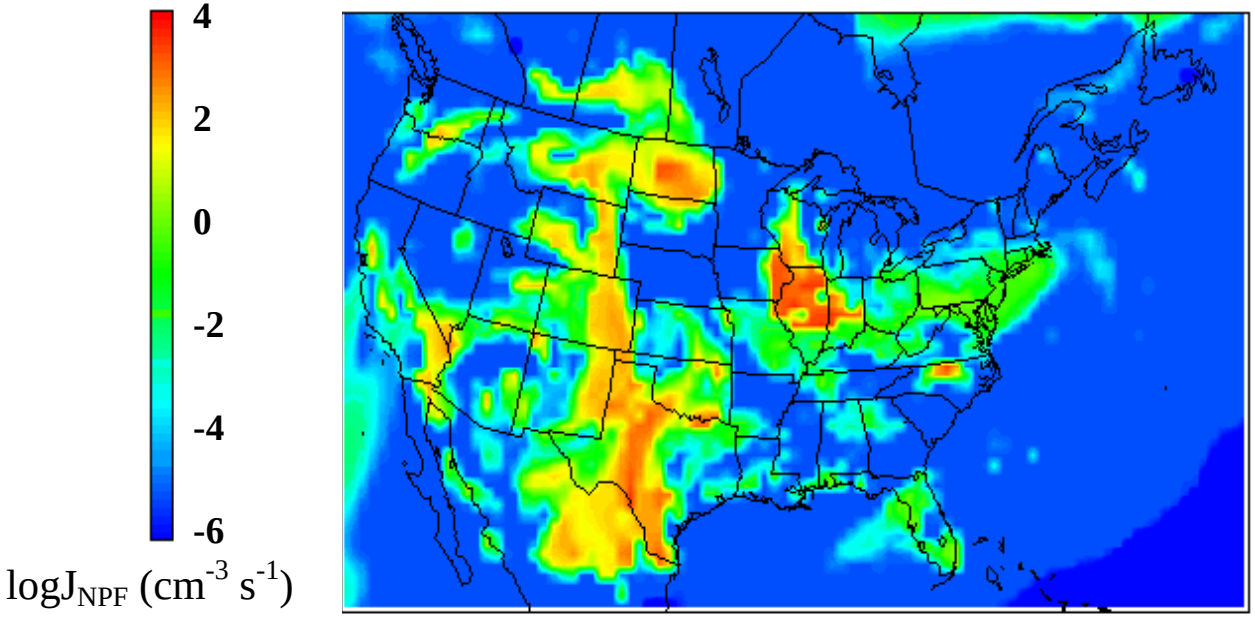
| Model Variants | Parameterizations Included | | | |
|---|---|---|---|---|
| | | NIIR | *F* | |
| 1 | N | + | + | |
| 2 | N | | + | |
| 3 | N | + | | |
| 4 | N | | | |
| 5 | V | + | + | |
| 6 | V | | + | |
| 7 | V | + | | |
| 8 | JVM | + | + | |
| 9 | N | + | + | $d_{g,emis} \times 2$ |
| 10 | N | + | + | $d_{g,emis} \div 2$ |
| 11 | N | + | + | IMTR |
| 12 | EM | | | |

## Parameterizations

| | | | |
|---|---|---|---|
| JVM | *Jaecker-Voirol and Mirabel, 1989* | Binary $H_2SO_4$–$H_2O$ nucleation rate: *J* | *theory* |
| V | *Vehkamaki et al., 2002* | Binary $H_2SO_4$–$H_2O$ nucleation rate: *J* | *theory* |
| N | *Napari et al., 2002* | Ternary $H_2SO_4$–$H_2O$–$NH_3$ nucleation rate: *J* | *theory* |
| NIIR | *Turco et al., 1998* | Ion-ion recombination nucleation rate: *J* | *theory* |
| *F* | *Kerminen and Kulmala, 2002* | New particle formation rate: $J_{NPF} = F_{KK02}\, J$ | *theory* |
| **EM** | *Eisele and McMurry, 1997* | New particle formation rate: $J_{NPF} = K[H_2SO_4]^2$ | *empirical* |
| **IMTR** | Intermodal Transfer | $F_{IMTR} = \left( \frac{D_{g,ATKN} - D_{g,NPF}}{D_{g,ACC} - D_{g,NPF}} \right)^p \quad (1 \le p \le 6)$ | |

# MODEL OVERVIEW

- Community Multiscale Air Quality (CMAQ) Model v4.4 with aerosol treatment by the modal approach
- Spatial domain : North America (96×60, 60 km horizontal resolution) with 21 vertical levels extending to 10 hPa
- Simulation Period : July - August, 2004 (ICARTT campaign)




Snapshot of new particle formation rate at 15UTC on July 6 at 600m altitude

# REACTION MECHANISM


$$\frac{d[\mathrm{H_2SO_4}]}{dt} = k[\mathrm{OH}][\mathrm{SO_2}] - k_{\mathrm{CS}}[\mathrm{H_2SO_4}] - n \times J_{\mathrm{NPF}}([\mathrm{H_2SO_4}])$$

| | |
|---|---|
| $k$ | Reaction rate coefficient |
| $k_{\mathrm{CS}}$ | Condensational sink rate of sulfuric acid on existing particles |
| $\mathrm{H_2SO_4}$ | Sulfuric acid concentration in gas phase |
| $n$ | Number of molecules of sulfuric acid in the new particles |
| $J_{\mathrm{NPF}}$ | New particle formation rate |

## Solution methods

- Assume steady state $\mathrm{H_2SO_4}$ without new particle formation
- Assume steady state $\mathrm{H_2SO_4}$ with new particle formation
- Time-dependent $\mathrm{H_2SO_4}$ with new particle formation

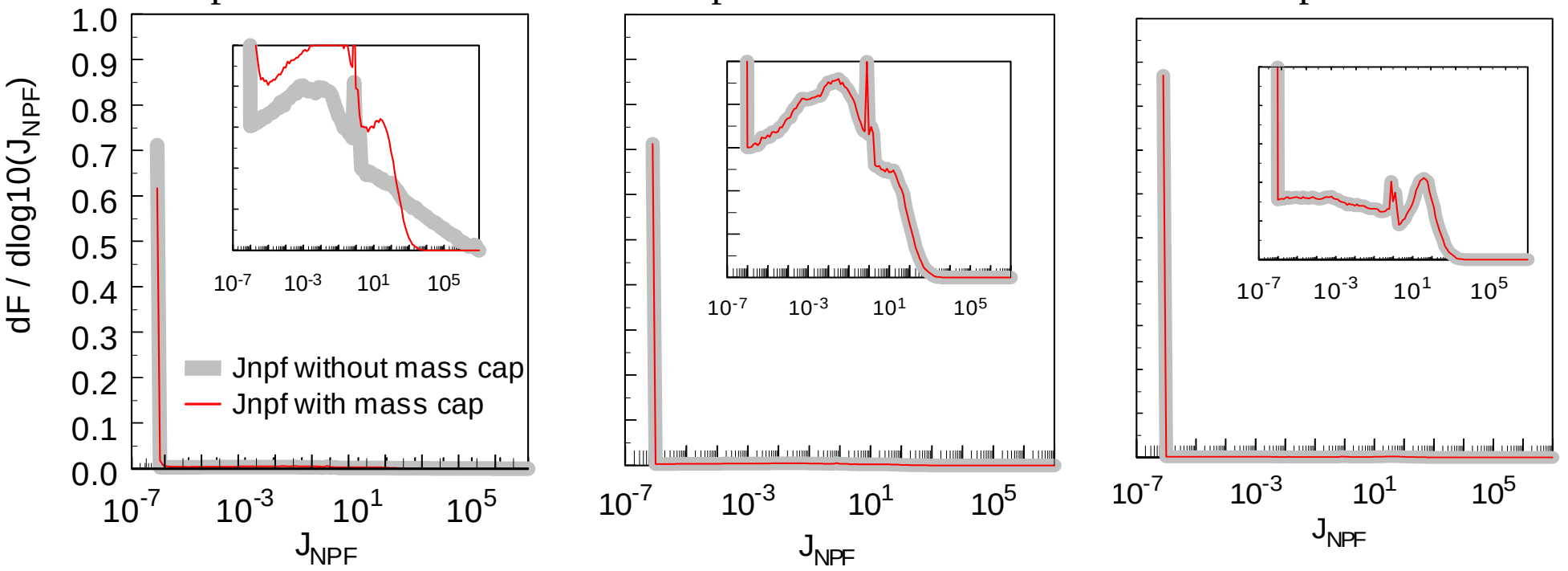
# SENSITIVITY OF NEW PARTICLE FORMATION TO SOLUTION METHOD

## Probability distribution function of $J_{NPF}$

Steady state $H_2SO_4$ without new particle formation

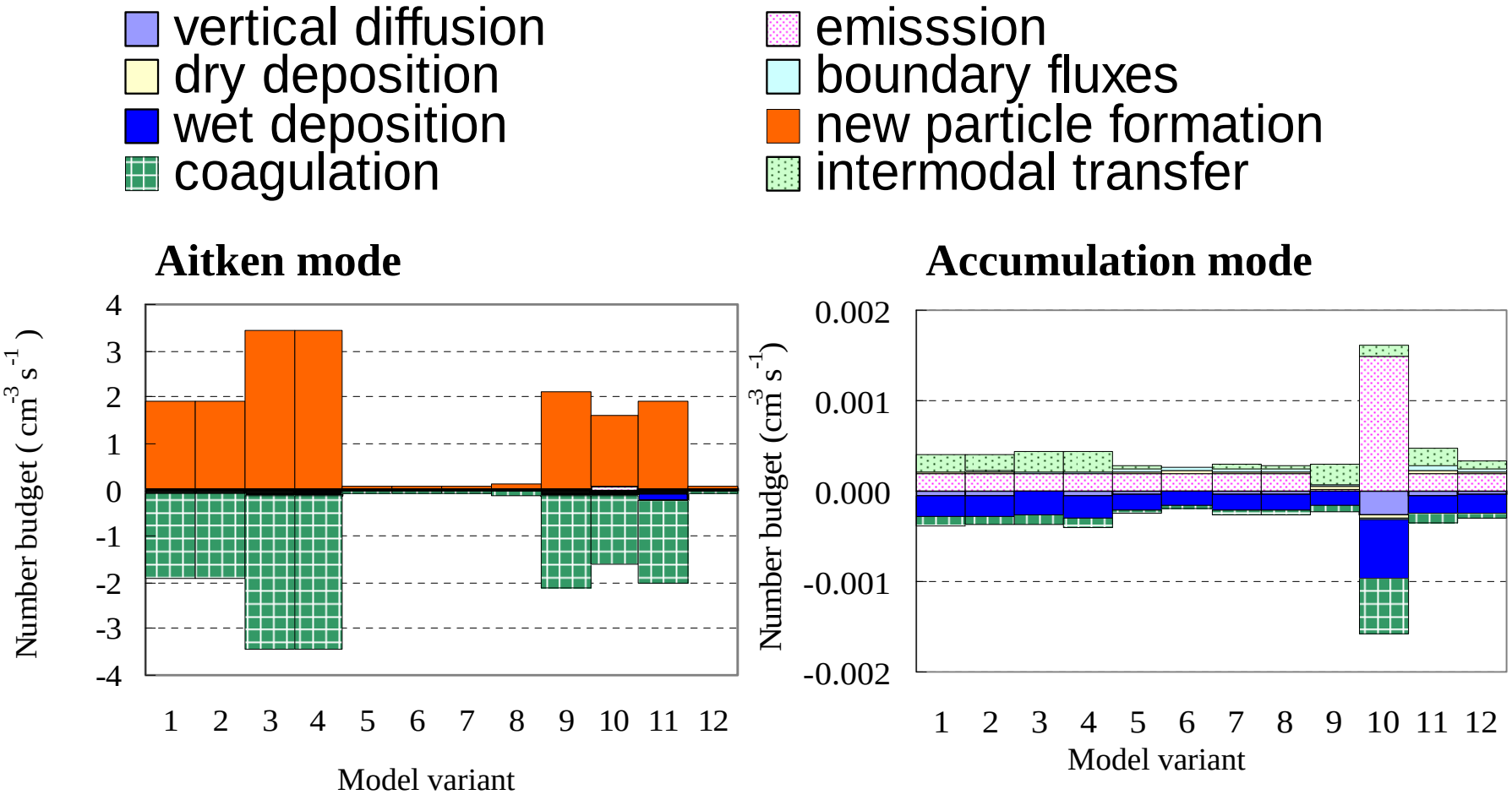
Steady state $H_2SO_4$ with new particle formation

Time-dependent $H_2SO_4$ with new particle formation




Time-dependent method accounts accurately for $H_2SO_4$ loss by condensational sink and new particle formation.

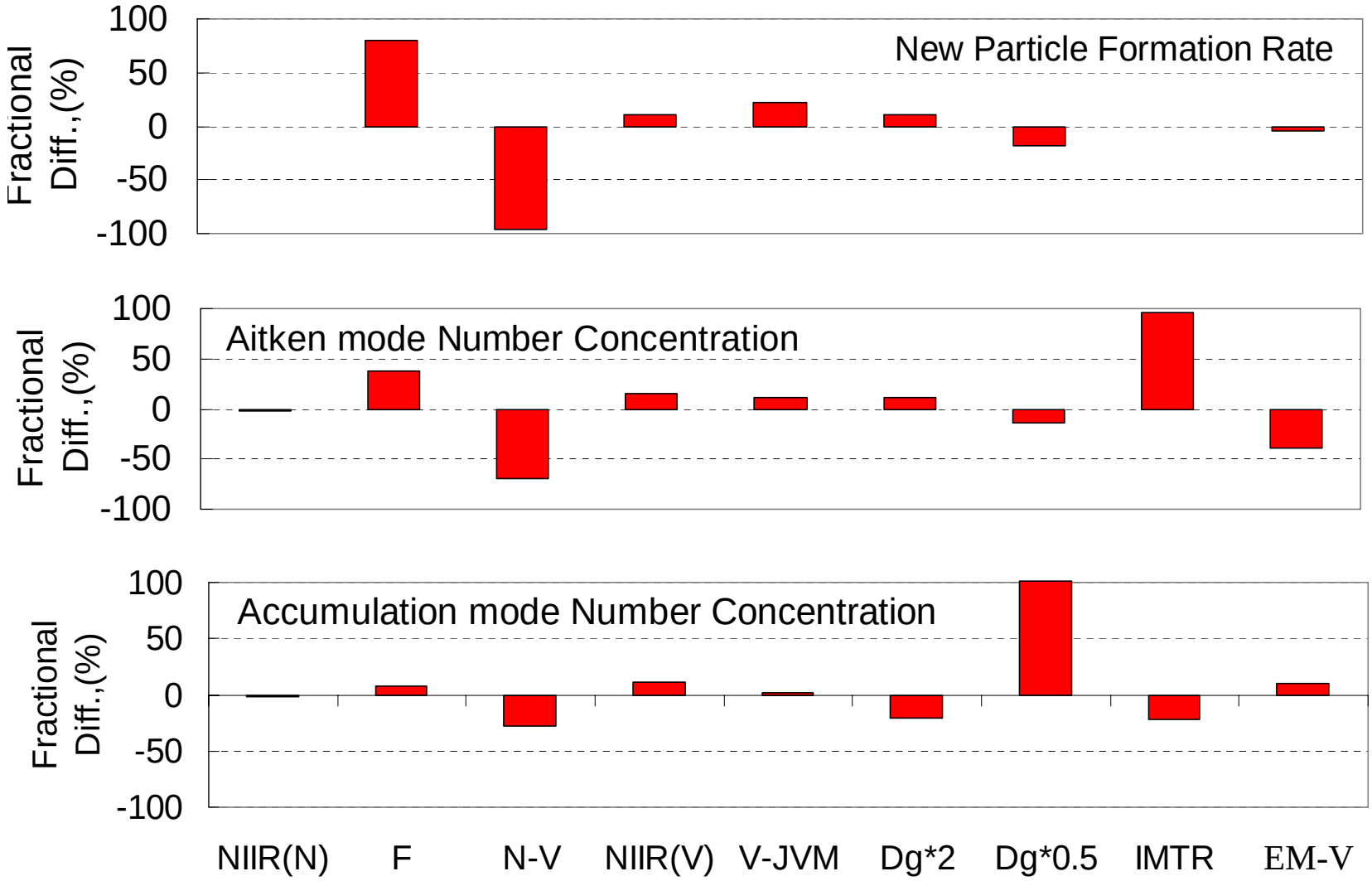
# NUMBER BUDGET ANALYSIS




In Aitken mode large variation of NPF is balanced largely by coagulation sink.

In accumulation mode primary emission is balanced largely by wet removal.

# KEY SENSITIVITIES




Aitken mode number concentration is most sensitive to nucleation mechanism and conversion process. Accumulation mode is sensitive to uncertainty of emitted particle size.

# CONCLUSIONS

- It is necessary to treat simultaneous time-dependent loss of $H_2SO_4$ by new particle formation and condensational sink to accurately compute $H_2SO_4$ concentration and new particle formation rate.
- The range of variation in average total **number concentration** over the model variants is **an order of magnitude smaller** than the range of variation in average **particle formation rates.**
- The uncertainty in average number concentration in the accumulation mode is largely due to uncertainty in the **conversion of mass emission rates to number emissions rates** rather than to uncertainty in new particle formation rates.