



# CMAQ Size Distributions in Pacific Northwest with Ternary Nucleation

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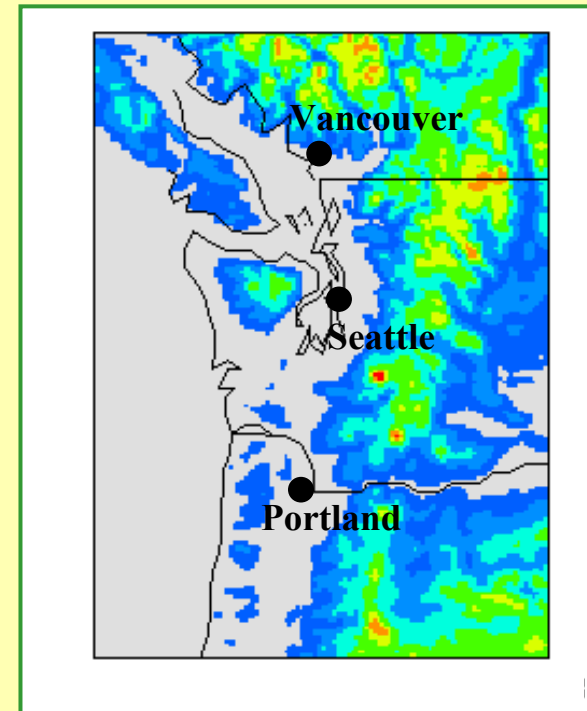
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# Overview

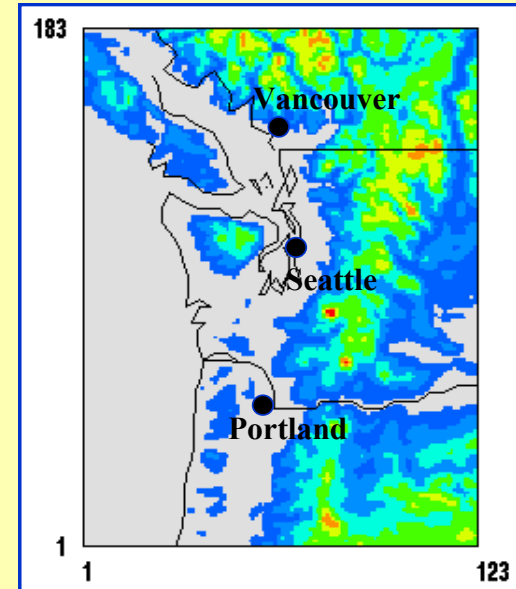
- **Goal**
  - More accurately model aerosol size distribution.
- **CMAQ v4.4 (September 2004)**
  - Aerosol treated as 3 log-normal modes
  - Kulmala (1998) binary  $\text{H}_2\text{SO}_4+\text{H}_2\text{O}$  nucleation
- **Poorly reproduces number concentrations despite otherwise average performance**
- **Ternary Nucleation for Pacific Northwest**
  - Test no nucleation, Kulmala binary, and Napari ternary nucleation
  - Compare to observations from field campaigns

CMAQ 4 km Domain



# CMAQ Simulation

- 4 km horizontal resolution
- 22 levels -- lowest layer is ~30 meters
- 4 day spin-up
  - 00 UTC 08/24 – 00 UTC 08/26 twice
- 3 day simulation
  - 00 UTC 08/26 – 00 UTC 08/29, 2001



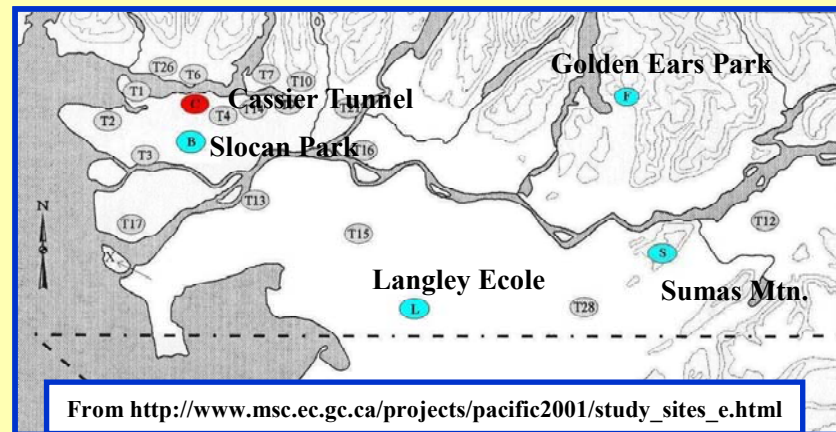
## Observational Datasets

### PNW 2001



From: <http://www.pnl.gov/pnw2001/>

### Pacific 2001

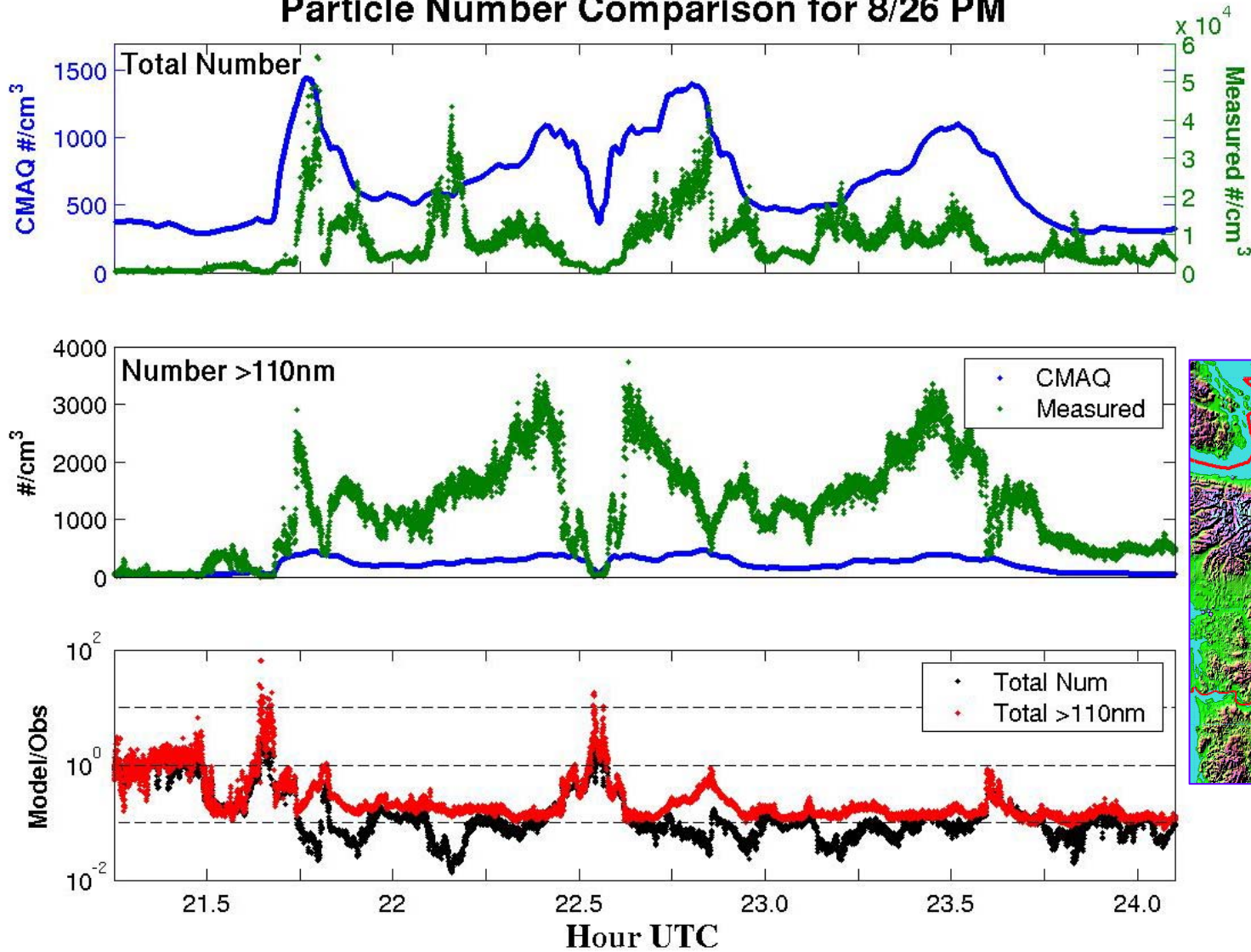


From [http://www.msc.ec.gc.ca/projects/pacific2001/study\\_sites\\_e.html](http://www.msc.ec.gc.ca/projects/pacific2001/study_sites_e.html)

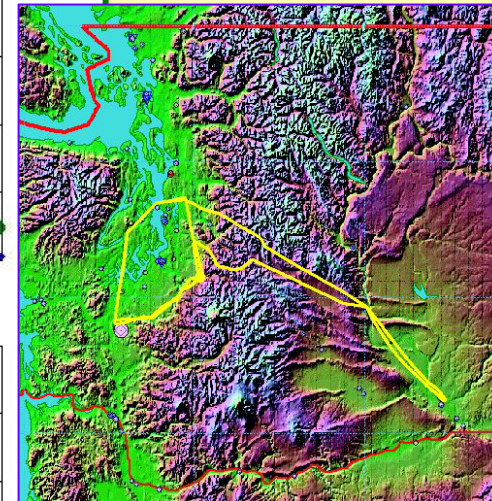


# Number Underprediction

## Particle Number Comparison for 8/26 PM



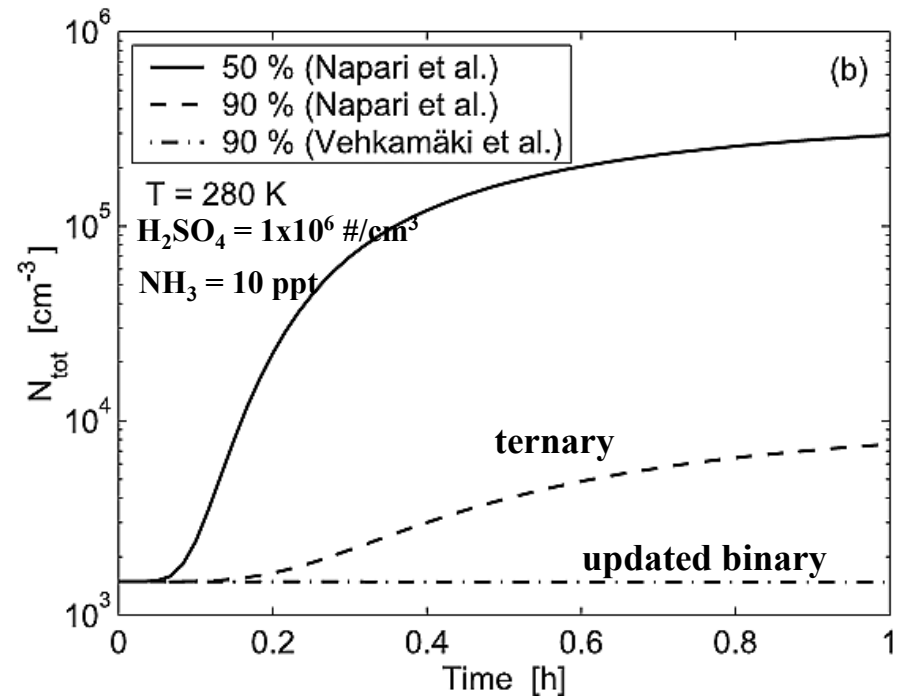
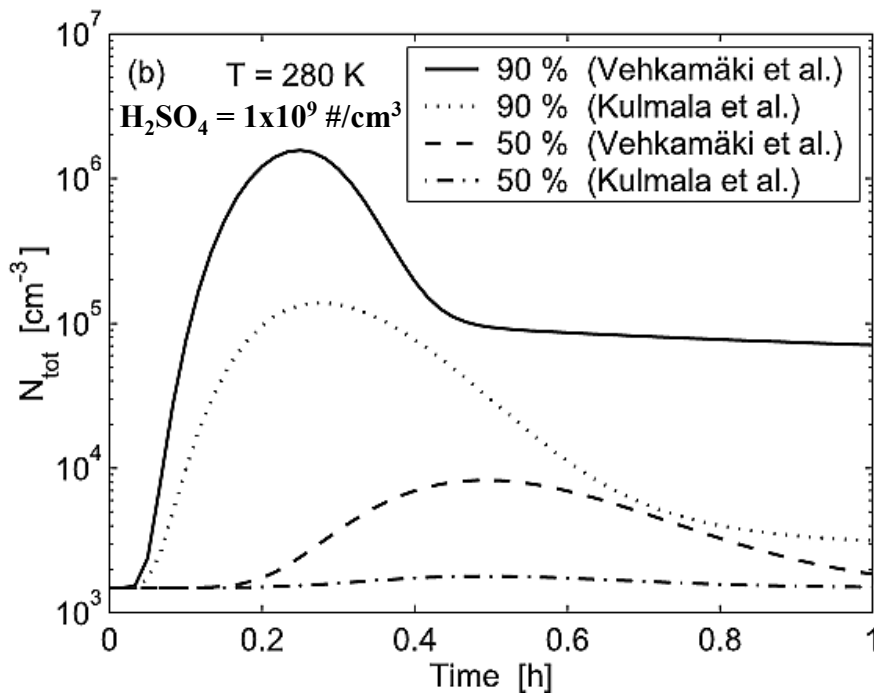
Flight track



Pacific Standard Time = UTC - 8

# Ternary Nucleation

Adapted from: Korhonen et al., 2003



- More current binary schemes produce more particles
- Ammonia is important component for sulfate-poor regions
- Enough  $\text{NH}_3$  for ternary nucleation to be significant

# Ternary Parameterization

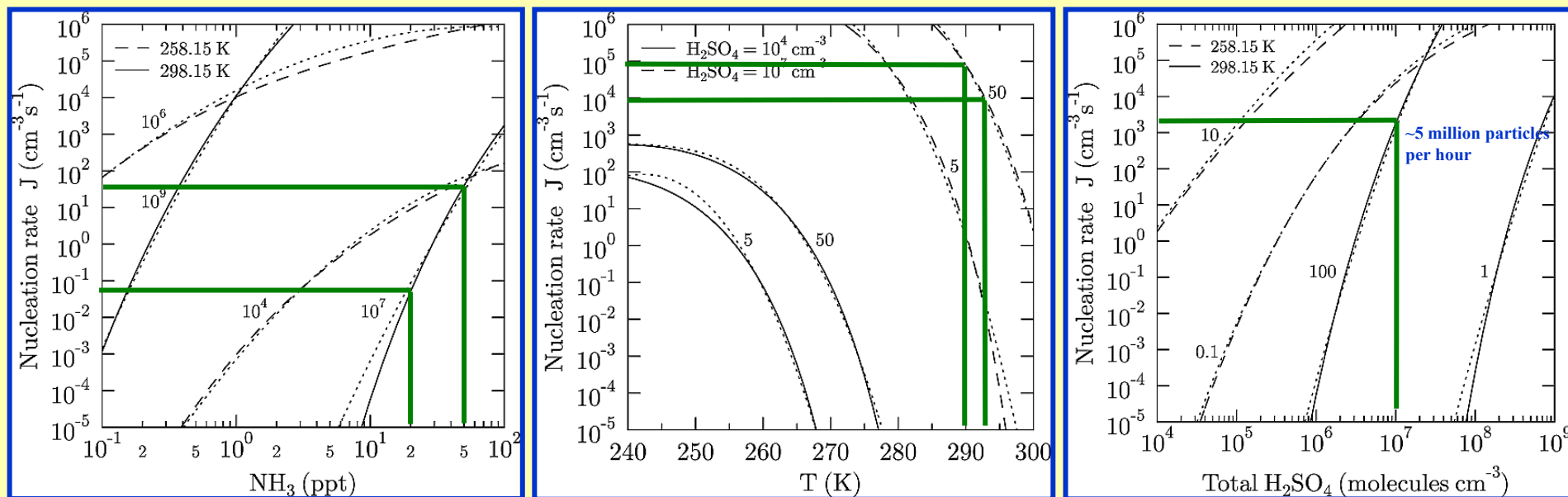
- Napari et al.

Napari, I., M. Noppel, H. Vehkamäki, and M. Kulmala, 2002: Parameterization of ternary nucleation rates for  $\text{H}_2\text{SO}_4\text{-NH}_3\text{-H}_2\text{O}$  vapors. *J. Geophys. Res.*, 107 (D19), 4381, doi:10.1029/2002JD002132.

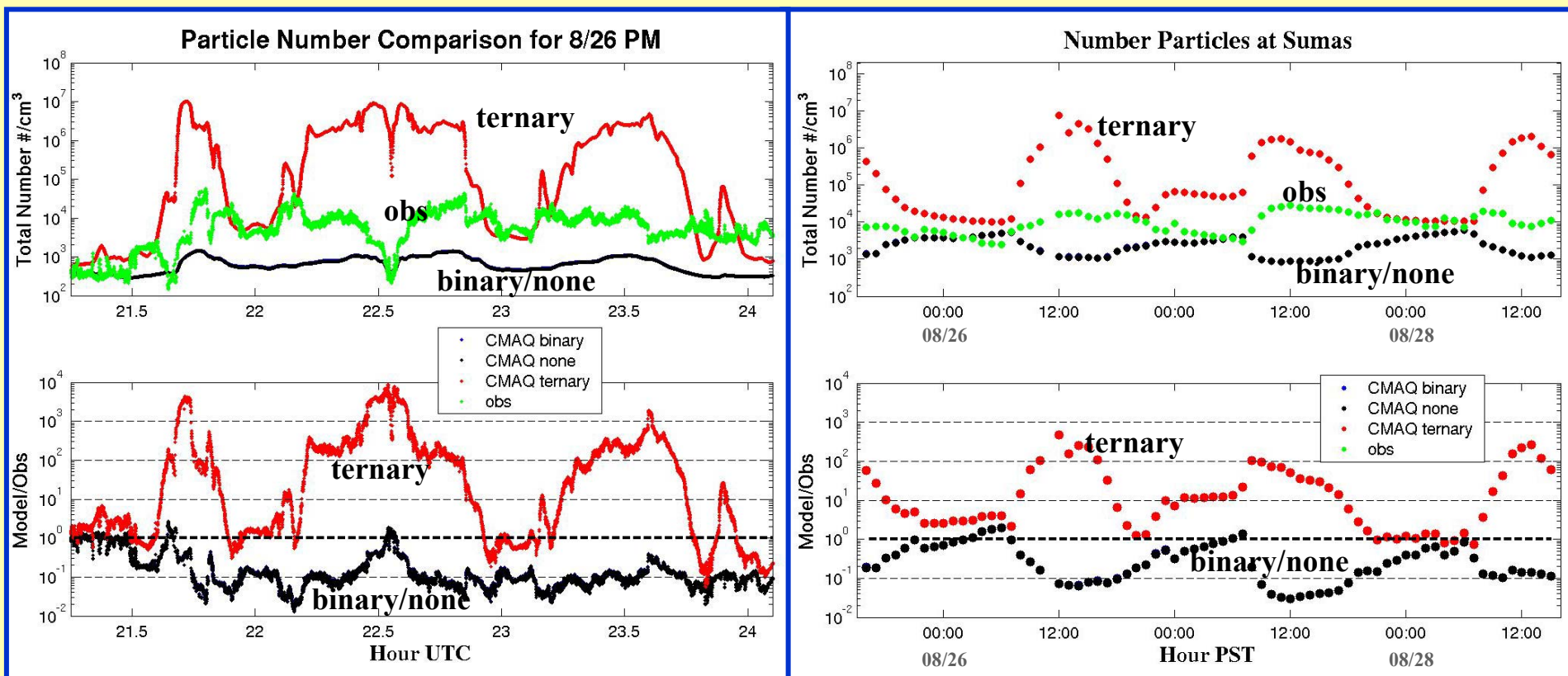
- Valid  $[\text{NH}_3]_{\text{g}}$  : 0.1 – 100 ppt

- Nucleation rate strong function of :

– Input  $[\text{NH}_3]_{\text{g}}$ , Temperature, Input  $[\text{H}_2\text{SO}_4]_{\text{g}}$



# Number Concentrations



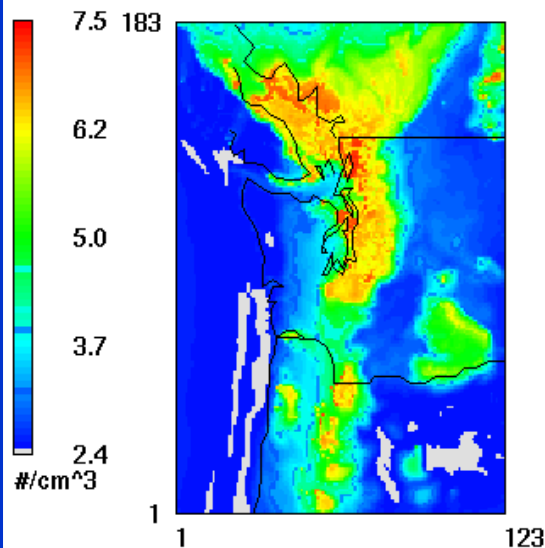
- No difference between binary nucleation and no nucleation
- Ternary nucleation overpredicts daytime urban number concentrations by about 3 orders of magnitude
- Ternary nucleation produces 5% more PM<sub>2.5</sub> than binary nucleation



# Spatial Distribution of Number Concentration

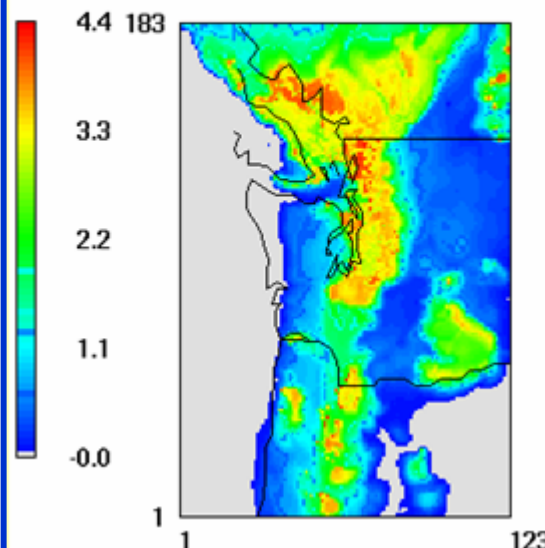
### Log of Ternary # Conc

August 26, 1PM PST  
~30 meters elevation



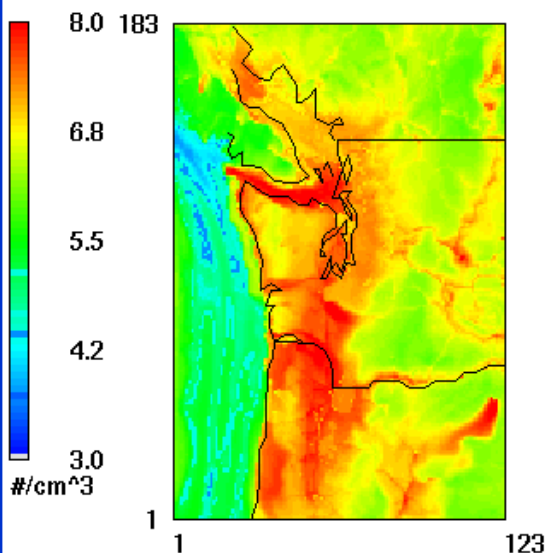
### Log of ternary/binary # conc

August 26, 1PM PST  
~30 meters elevation



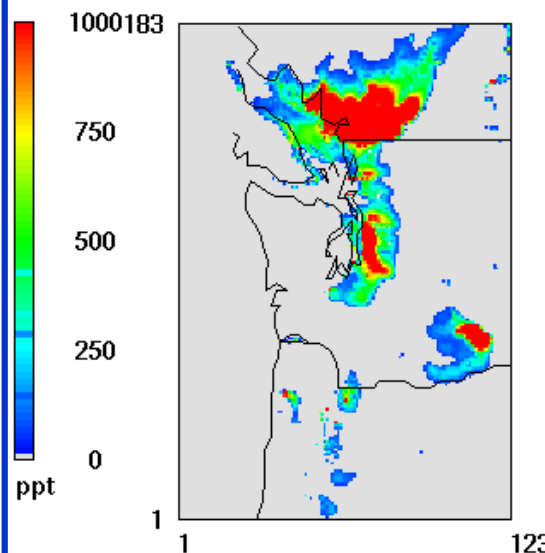
### Log of Sulfuric Acid Conc

August 26, 1PM PST  
~30 meters elevation



### Ammonia Concentration

August 26, 1PM PST  
~30 meters elevation

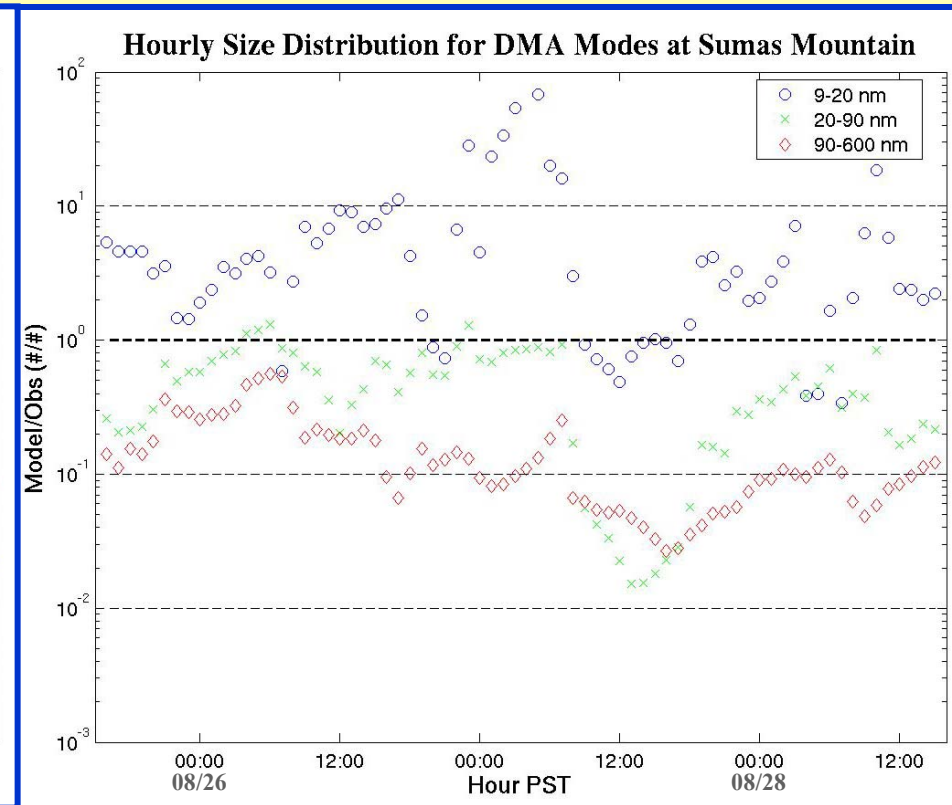
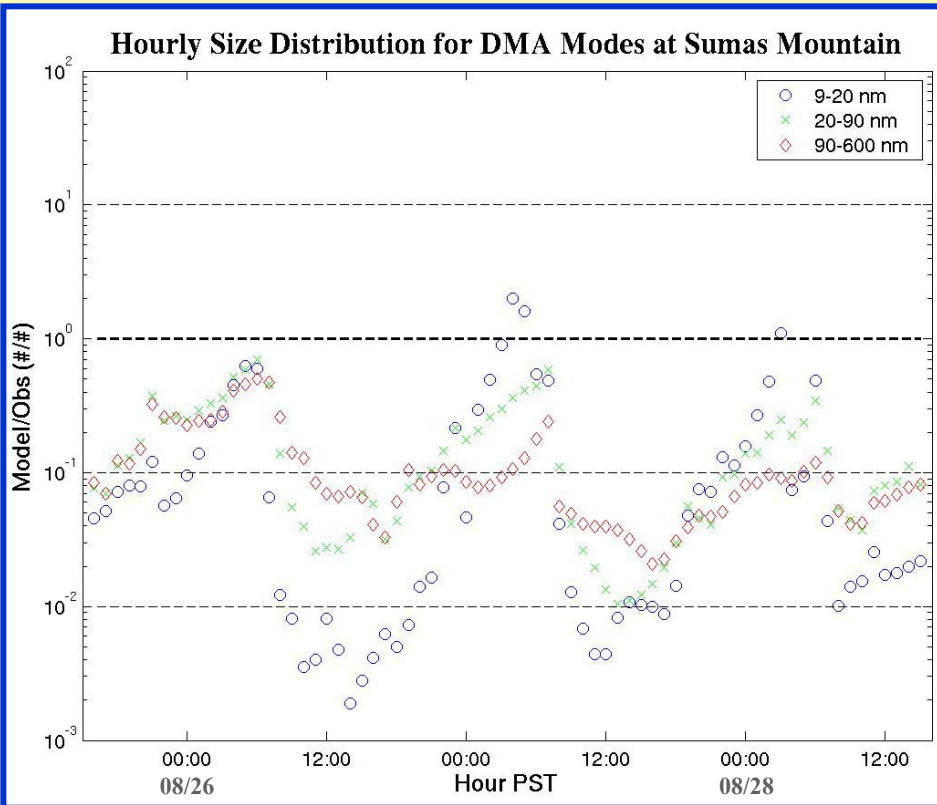




# Size Distributions

## Binary Nucleation

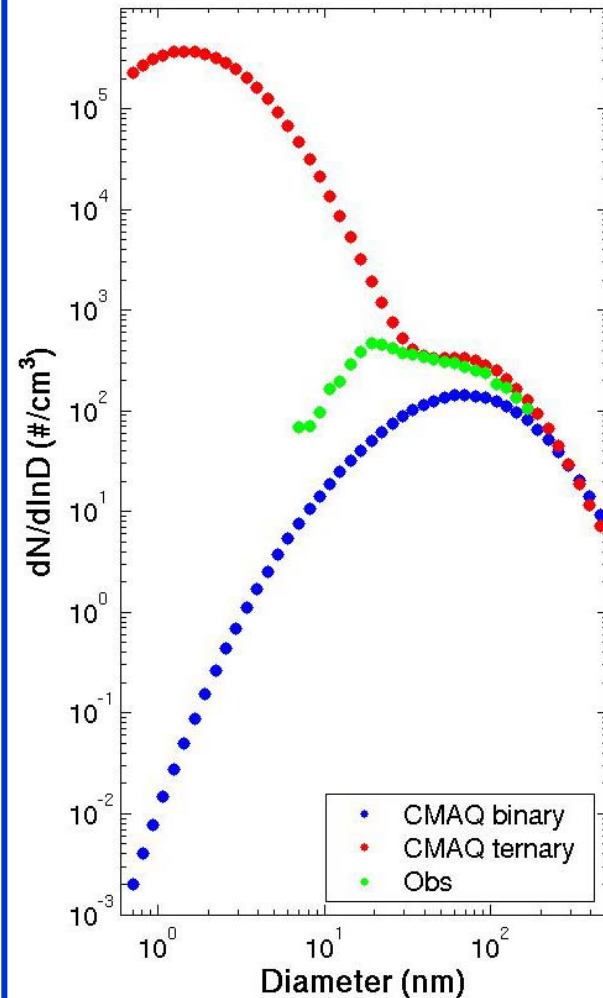
## Ternary Nucleation



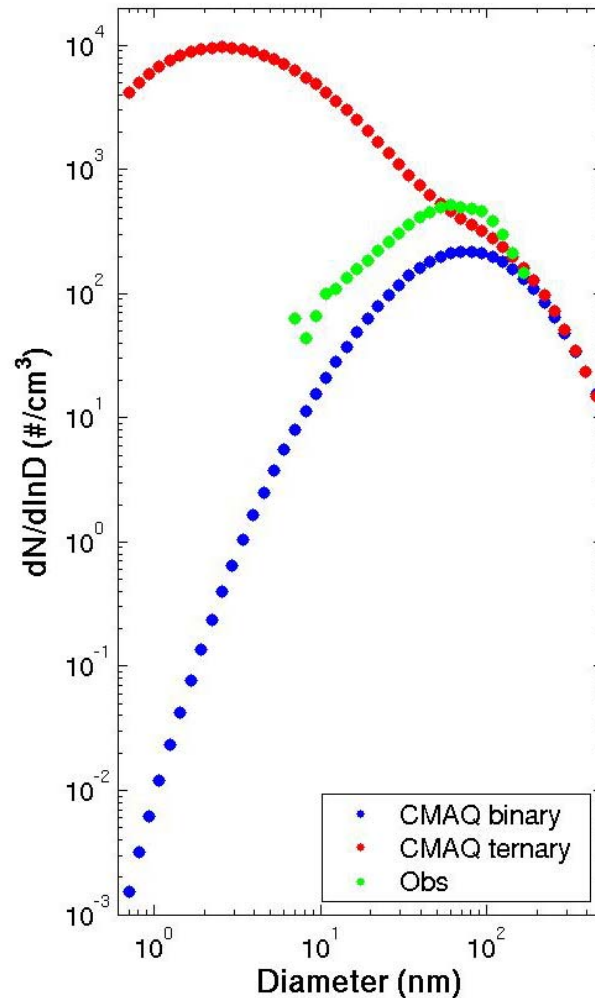
- Nucleation mode is overpredicted with ternary nucleation.
- Aitken mode is modeled better with ternary nucleation.
- No change in accumulation mode performance.

# Ternary Shifts i-mode to Small Sizes

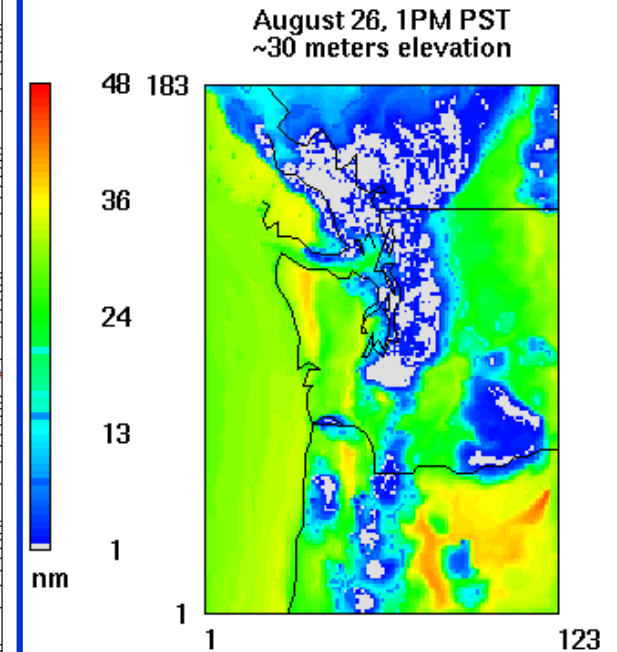
Daytime Langley # Distribution



Nighttime Langley # Distribution



i-Mode Median Diameter



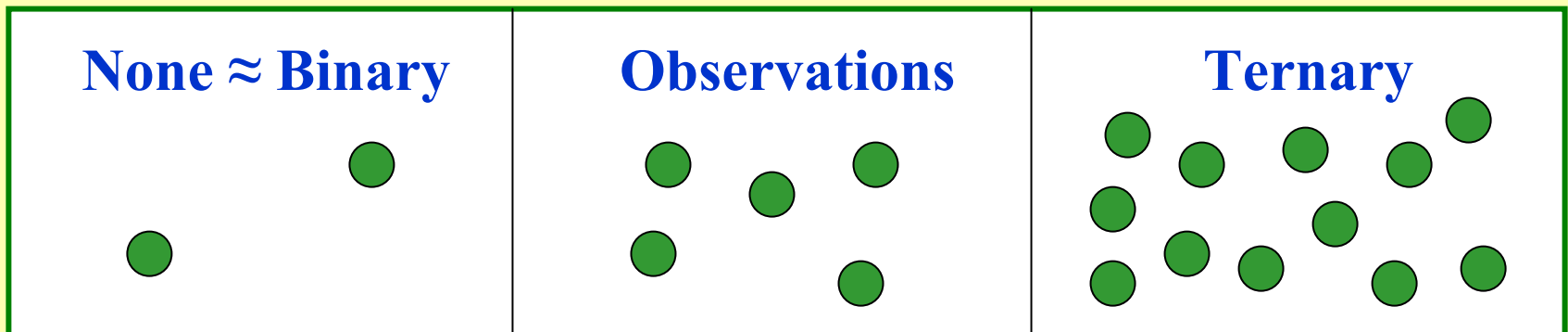
- Fresh particles dominate i-mode
- Effect greater during day, nearly absent in rural areas

# What is Going On?

- **Nucleation mode physics**
  - Not captured with current 3-mode model
  - Near-molecular dynamics and growth not accounted for
- **Ternary Nucleation theory**
  - Effect of ammonium bisulfate (Anttila et al., in press)
  - Not a solved problem

# Conclusions

- **CMAQ v4.4 binary nucleation produces very few particles in this simulation**
- **Ternary nucleation**
  - Produces urban concentrations in the millions instead of tens of thousands
  - Shifts the i-mode to 1-2 nm
  - Increases  $PM_{2.5}$  by  $\sim 5\%$
- **Highlights importance of the “h” mode.**
- **Nucleation theory is still incomplete**





# Hourly Size Distributions for 08/2001

