CMAQ Size Distributions in Pacific Northwest with Ternary Nucleation

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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 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

Pacific 2001

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

From: http://www.msc.ec.gc.ca/projects/pacific2001/study_sites_e.html
Number Underprediction

Particle Number Comparison for 8/26 PM

Total Number

Number >110nm

Model/Obs

Pacific Standard Time = UTC - 8
Ternary Nucleation

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

Adapted from: Korhonen et al., 2003
Ternary Parameterization

- **Napari et al.**
  

- **Valid [NH$_3$]$_g$ : 0.1 – 100 ppt**

- **Nucleation rate strong function of:**
  - Input [NH$_3$]$_g$, Temperature, Input [H$_2$SO$_4$]$_g$
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$_{2.5}$ 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
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

- 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 ~ 5%
• Highlights importance of the “h” mode.
• Nucleation theory is still incomplete

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<thead>
<tr>
<th>None ≈ Binary</th>
<th>Observations</th>
<th>Ternary</th>
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<tr>
<td>None</td>
<td>Observations</td>
<td>Ternary</td>
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