SENSITIVITY OF AEROSOL PARTICLE NUMBER CONCENTRATION AND SIZE TO NEW PARTICLE FORMATION MECHANISM AND PRIMARY EMISSIONS IN A CONTINENTAL SCALE CHEMICAL TRANSPORT MODEL

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MOTIVATION

SIZE AND NUMBER CONCENTRATION MATTER

- Direct effects: light scattering and absorption, RH dependence
- Indirect effects: CCN as function of supersaturation
- Aerosol removal processes: dry and wet deposition
- Aerosol influences on precipitation
- Effects on new particle formation and subsequent aerosol dynamics

THE MODEL

THIS STUDY

- Examines the sensitivity of observable aerosol properties to formulations of new particle formation and properties of primary emitted particles.
- Examines four theoretical formulations and one empirical formulation of new particle formation.
- Examines the effect of doubling or halving diameters of primary particles with mass emissions held constant (number concentration decreased or increased eightfold) on particle concentrations, new particle formation, and aerosol dynamics.
- Presents results for a one-month simulation (July 2004) using a continental-scale model for the United States.
- Examines specific situations using a higher resolution embedded model.
- Compares modeled aerosol properties with measurements.

KEY MODEL ATTRIBUTES

Model platform: Community Multiscale Air Quality model (CMAQ)

Key processes: Advection, gas phase chemistry, aqueous sulfate chemistry, aerosol thermodynamics, aerosol microphysics, redistribution of aerosols in convective clouds, and dry and wet removal.

Gas phase chemistry: SAPRC99. 72 species, 214 reactions: inorganics (NO_x, O_3, H_2O_2, CO, SO_2 ...), radicals (OH, HO_2, organic peroxy radicals ...) organics (PANs, lumped alkenes, aromatics, and olefins, isoprene, terpenes ...).

Nucleation: sulfuric acid, water, and ammonia. *Sensitivity to nucleation mechanism examined.*

Secondary organic aerosol formation from anthropogenic and biogenic precursors.

New particle criterion: Growth to 3 nm diameter by condensation of sulfuric acid, water, and organics. New particles enter the Aitken mode.

Aerosol microphysics: Modal representation with three modes (Aitken, accumulation, and coarse) and three moments per mode.

Model domain: Continental US at 60 km resolution; 21 vertical levels to 10 hPa; 15 min time step. Northeast US: nested domain with 12 km resolution.

Meteorological fields (temperature, relative humidity, wind speed and direction, cloud/rain properties, and turbulent fluxes): MM5.

Anthropogenic emissions: EPA 1999 National Emission Inventory updated for a typical summer day of 2004.

Hourly gridded emissions: Sparse Matrix Operator Kernel Emissions (SMOKE).

Primary emitted species: NO, NO_2, VOC, CO, SO_2, NH_3, 41 speciated VOCs, 5 PM2.5 aerosol species (sulfate, nitrate, OC, EC, and unspecified), PM10.

Primary particle emissions: 99.9% of total emitted mass into the accumulation mode and 0.1% into the Aitken mode. Geometric mean diameter in Aitken mode 30 nm; in accumulation mode 300 nm. *Sensitivity to primary particle size is examined.*

Biogenic emissions: Biogenic Emissions Inventory System (BEIS 3.11).

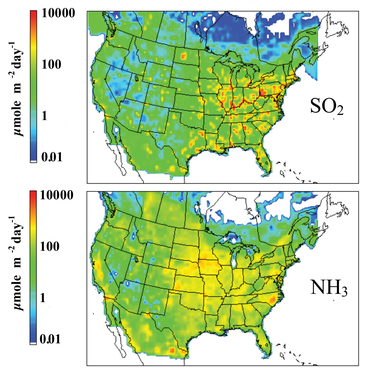
Biomass burning emissions: Not included.

Comparison with observations: ICARTT, July-August 2004.

EMISSIONS

EMISSIONS OF SO_2 AND NH_3

Important as nucleation precursors



SO_2 >> NH_3 in eastern US but NH_3 >> SO_2 in midwest and west.

EXPERIMENTS

MODEL VARIANTS

Base case (no NPF) plus 8 variants

Model Variant	Nucleation mechanism	NPF conversion factor	$D_{g,emission}$ / $D_{g,emission,BASE}$
BASE	No NPF		1
BHNV	BHN (VEH)	KK	1
BHNJ	BHN (JVM)	KK	1
NIIR	NIIR	KK	1
EM97	EM97	NA	1
THN	THN	KK	1
THN + NIIR	THN + NIIR	KK	1
$Dg \times 2$	THN + NIIR	KK	2
$Dg \div 2$	THN + NIIR	KK	1/2

THN Ternary homogeneous nucleation of sulfuric acid, water and ammonia.

NIIR Nucleation by ion-ion recombination.

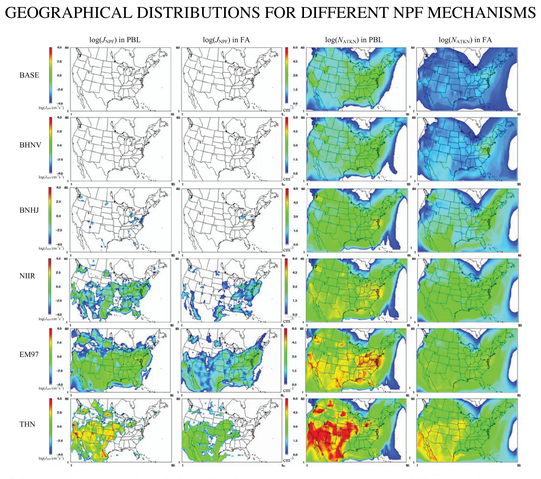
BHN Binary homogeneous nucleation of sulfuric acid and water with rates given by Vehkamäki (2002, VEH) or Jaecker-Voirol and Mirabel (1989, JVM).

EM97 Empirical rate expression for NPF of Eisele and McMurry (1997).

KK Fraction of nucleation events that attain diameter of 3 nm, (Kerminen and Kulmala, 2002).

FINDINGS

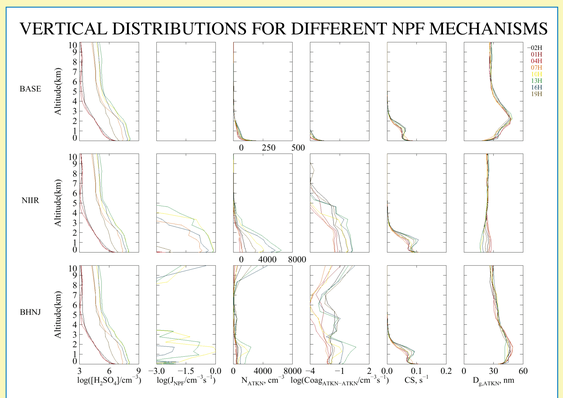
GEOGRAPHICAL DISTRIBUTIONS FOR DIFFERENT NPF MECHANISMS



Modeled geographical distributions depend strongly on NPF mechanism.

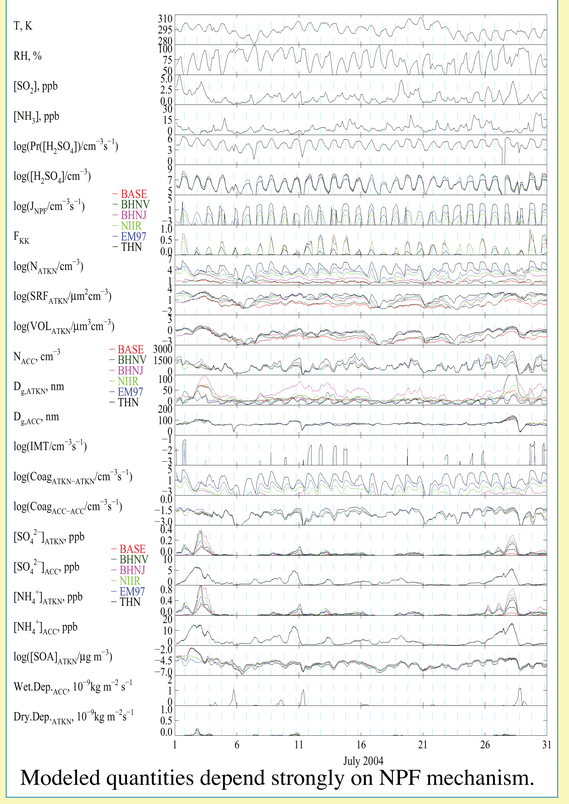
FINDINGS

VERTICAL DISTRIBUTIONS FOR DIFFERENT NPF MECHANISMS



Modeled vertical distributions depend strongly on NPF mechanism.

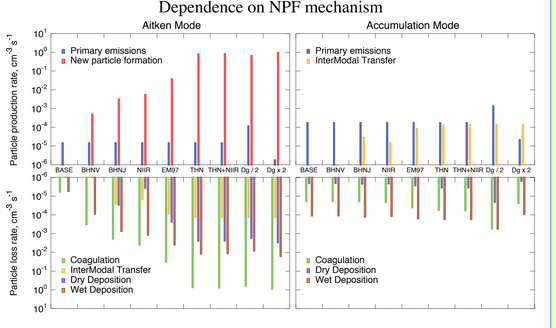
TIME SERIES FOR DIFFERENT NPF MECHANISMS



Modeled quantities depend strongly on NPF mechanism.

BUDGET OF PARTICLE NUMBER CONCENTRATION

Dependence on NPF mechanism

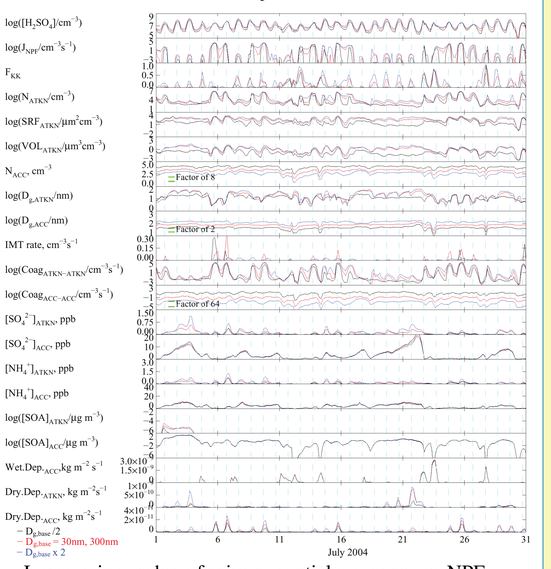


Increased NPF is compensated mainly by increased coagulation.

FINDINGS

SENSITIVITY TO SIZE OF PRIMARY EMISSIONS

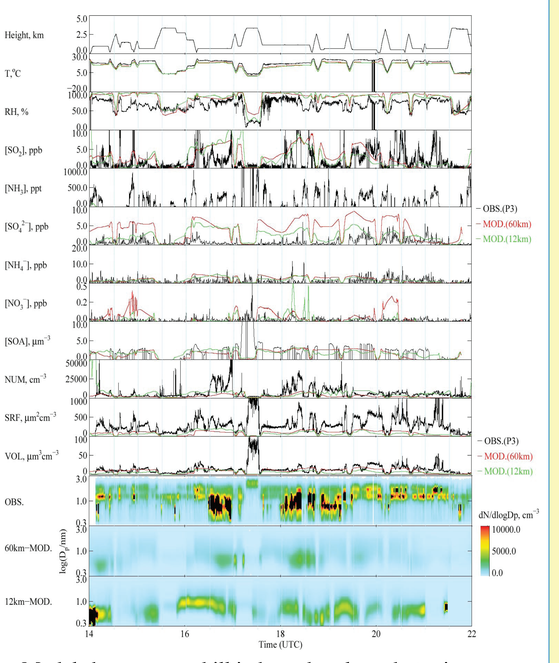
Factor of 2 decrease in diameter corresponds to factor of 8 increase in number



Increase in number of primary particles suppresses NPF.

COMPARISON WITH OBSERVATIONS

Measurements on NOAA P-3 during ICARTT



Model shows some skill in boundary layer but misses fine structure even at 12 km resolution.

Model misses much aerosol in free troposphere.

KEY CONCLUSIONS

- NPF rate and Aitken mode number concentration are highly sensitive to NPF mechanism and NH_3.
- Number concentration and NPF rate are highly sensitive to emitted particle size.
- Measurements are needed to constrain NPF rate and dependence on controlling variables.
- Model shows skill in particle volume and aerosol species concentrations.
- Continental scale model is highly sensitive to emissions outside model domain, especially in free troposphere.