Modeling Aerosol Processes in the DOE Atmospheric Science Program

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Objective

Develop and evaluate aerosol microphysical models (modules) representing the processes governing the chemically-resolved particle size distribution

Approaches

- Examine modal / moment / sectional representations of the size PDF
- Use 1-D / 2-D / 3-D gridded host models for aerosol module evaluation
- Explore (novel) methods for incorporating field measurements in host models

Applications

- Model summer 2004, NE US, probable focus on SOA [2005]
- Model ASP campaigns and examine long-range aerosol influence [2006-07]

Host Chemical Transport Models

CMAQv4.4 [Community Multiscale Air Quality Modeling System – EPA]

- Urban-to-regional with 1-way nested gridding
- Well-suited for interchanging and comparing alternate modules while retaining the same meteorology
- Well-suited for configuration in 1-D/2-D to provide departure points for developing ‘observation-based’ (plume) models
- Current aerosol module options: AERO3, MADRID (1, 1a, 2)

GChM-O [Global Chemistry Model driven by Observation-derived meteorology – BNL]

- Regional-to-global with 1-way nested gridding
- Suitable for examining long-range aerosol influence
- Original version: sulfur-only with tagging by geographical source region
- Present version: includes sub-model with RADM2 + RADM + AERO3

Current Aerosol Modules

	AERO3	MADRID 1, 1a	MADRID 2
Size distribution	modal (3 modes) number PDF	sectional (2, 8 sections) mass PDF	sectional (2 sections) mass PDF
Nucleation	absolute rate	relative rate	relative rate
Condensational growth	modal	moving-center	not treated
Coagulation	modal	not treated	not treated
Gas-particle mass transfer	full equilibrium	CIT equilibrium or CMU hybrid	CIT equilibrium or CMU hybrid
Inorganics	SO_4^{2-}, NH_4^+, NO_3^-	SO_4^{2-}, NH_4^+, NO_3^-, Na^+, Cl^-	SO_4^{2-}, NH_4^+, NO_3^-, Na^+, Cl^-
Secondary Organic Aerosol	"parameterized" (Schell et al. 2001) 2 predicted species: 1 anthropogenic 1 biogenic	"parameterized" (Odum et al . 1997, Griffin et al. 1999) 38 predicted surrogate species: 4 anthropogenic 34 biogenic	"first principles" (Pun et al. 2002) hydrophilic / hydrophobic 42 predicted explicit species: 31 anthropogenic 11 biogenic
Gas-phase chemistry	CBM-IV (46, 96) RADM2 (75, 200) SAPRC-99 (79, 214)	CBM-IV (92, 117) RADM2 (121, 221)	CACM (191, 361) (Griffin et al. 2002)
Aqueous chemistry	RADM (33, 23)	RADM (33, 23) CMU (55, 133)	CMU (55, 133)
Heterogeneous chemistry	not treated	$HO_2/NO_2/NO_3/N_2O_5$ on surfaces N_2O_5 in droplets (Jacob 2000)	$HO_2/NO_2/NO_3/N_2O_5$ on surfaces N_2O_5 in droplets (Jacob 2000)
Features to be added	alternate nucleation schemes	coagulation increased size resolution	coagulation increased size resolution

New Aerosol Module: AERO-QMOM

- Extends the multi-modal concept of AERO3 to generally-mixed aerosols
- Closes the moment equations without assumed form of the PDF (QMOM)
- Uses Principal Components Analysis for multivariate quadratures (PCA-QMOM)

Strengths

- Computational efficiency
- Freedom from numerical diffusion in particle size space
- Efficient evaluation of aerosol optical properties in coupled meteorology-chemistry models via quadrature integral evaluation
- Computationally feasible treatment of generally-mixed aerosols

Limitations

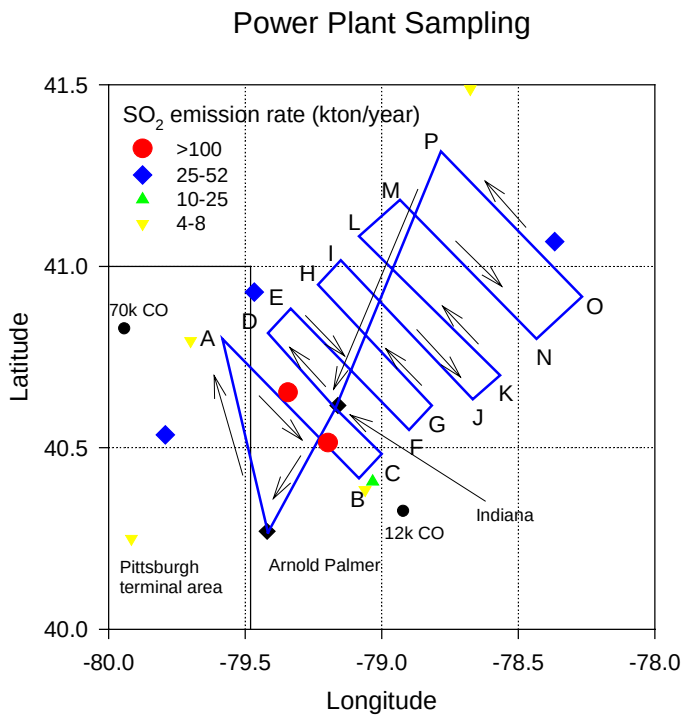
- Limited size-resolved information
- Loss of information (sometimes required moment interrelationships) during transport
  - Assessment of problem needed

Unknowns

- Treatment of size-resolved multicomponent aerosols when merging the QMOM dynamics with equilibrium and gas-particle partitioning modules

1-D / 2-D 'Plume' Modeling

Explore closer synthesis of models with field measurements at campaign locations



Develop 'observation-based' models that constrain some quantities normally predicted in 'emission-based' models:

- Constrain plume dimensions, gas-phase concentrations (SO_2, VOCs) to observations
- Explore analogs for aerosols of the constrained steady-state calculations used to determine O_3 production rates
- Examine the evolution of the chemically-resolved aerosol size distribution
- Build models within the CMAQ system with additional components

What We Need

NE US, Summer 2004 / Mexico City, Winter 2006

- Gridded meteorology (MM5, WRF-CHEM also for Mexico City)
- Gridded emissions (gas, particulate)
    - Chemically-speciated organic particulate emissions
- Surface data: aerosols, trace gases, meteorology (sounders, rawinsondes)
    - From Indiana, PA [2004]
- Data from all G-1 participants

What We Can Provide [Deliverables]

- Gridded (3-D) fields of chemically-resolved aerosol size distributions
- All of our models / modules

? Assessment of moment transport problem