Effects of Cloud-Aerosol-Chemistry Interactions on Aerosol and Cloud Optical Depths in Western Pennsylvania During August 2004

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Motivation

- estimate the impact of aerosol radiative forcing on local and regional meteorological processes and better understand the uncertainties associated with:
  - direct effect: aerosol-radiation feedbacks (scattering and absorption)
  - indirect effect: aerosol-cloud feedbacks

- aerosol-radiation feedbacks implemented last year

size and number distribution, composition, aerosol water → *refractive indices* → *Mie theory* → *3-D τ_λ, ω_o, and g* → *scattering and absorption of shortwave radiation*

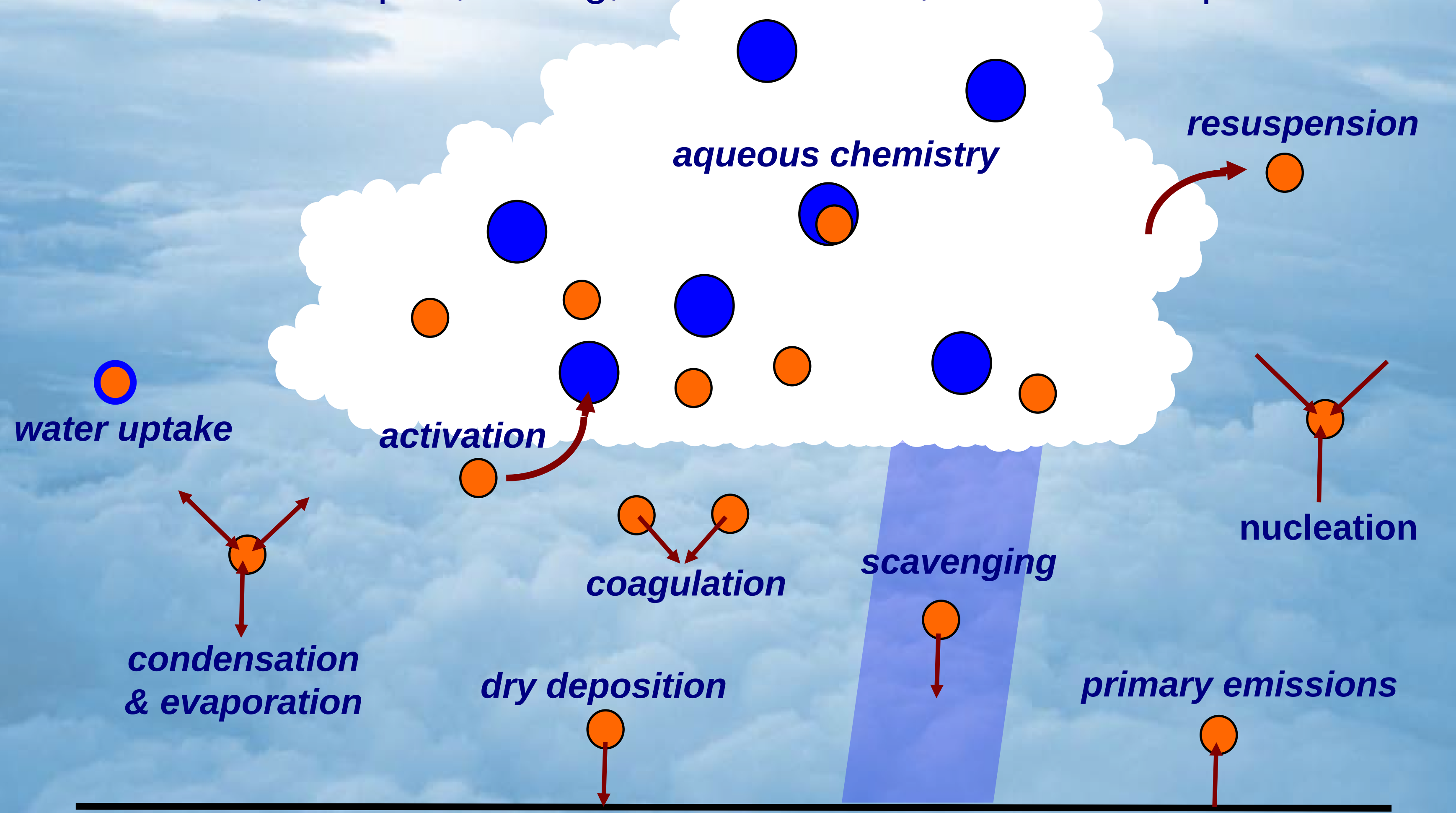
- aerosol-cloud feedbacks implemented this year

aerosol number → *aerosol activation* → *prognostic cloud droplet number, aqueous chemistry* → *cloud albedo, precipitation, cloud lifetime* → *wet removal*

  - first aerosol indirect effect (change in cloud albedo)
  - second aerosol indirect effect (change in precipitation)
  - coupled with MOSAIC aerosol model only

Aerosol Processes

- simulate life-cycle of aerosols in WRF-chem framework that includes emissions, transport, mixing, transformation, and removal processes



Prognostic Cloud Droplet Number

- converted *Lin et al.* microphysics scheme (*mp_physics=2*) to a two-moment treatment (mass & number)

$$\frac{\partial N_k}{\partial t} = -(V \bullet \nabla N)_k + D_k - C_k - E_k + S_k$$

N_k - grid cell mean droplet number mixing ratio in layer k
D_k - vertical diffusion
C_k - droplet loss due to collision/coalescence & collection
E_k - droplet loss due to evaporation
S_k - droplet source due to nucleation

- cloud droplet number source determined by aerosol activation (for meteorology-only runs a prescribed aerosol size distribution is used)
- droplet number and cloud water mixing ratio used to compute effective cloud-particle size for computing cloud optical depth in Goddard shortwave radiation scheme (*ra_sw_physics=2*)

New Variables in WRF-chem

- new variables added to Registry:
  - ***qndrop***, droplet number mixing ratio
  - ***qndropsource***, droplet number source
  - ***qlsink***, cloud water fractional removal rate needed for wet scavenging
  - ***ccn1*** - ***ccn6***, CCN concentration at S of 0.02, 0.05, 0.1, 0.2, 0.5, and 1.0%

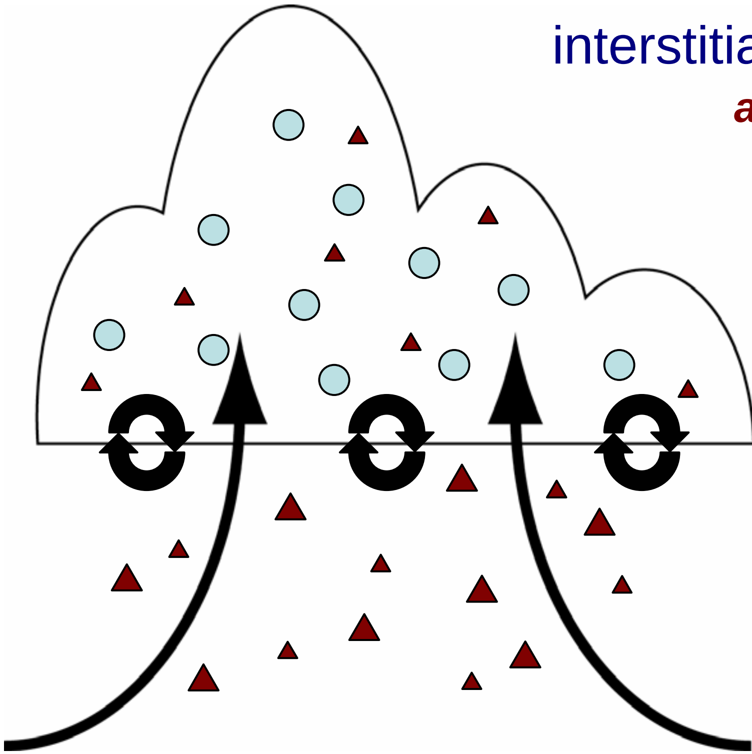
- interstitial and cloud-borne aerosol species
  - ***so4_a01*** = sulfate bin #1 and ***so4_cw01*** = cloud-borne sulfate bin #1
  - … similarly for other composition and size bins

- employ generalized pointer array, ***massptr_aer(c,s,t,p)***
  - **c** =composition, **s** =size bin, **t** =type, **p** =phase (interstitial, cloud, ice, rain)
  - <u>problem</u>: as the complexity of aerosol mechanisms increase, uniquely identifying each variable within the *chem* field array becomes cumbersome
  - <u>solution</u>: a pointer array that groups the aerosol variables by specific characteristics so that looping overall species is now simple
  - would be useful to have pointer array at Registry level

Aerosol Activation and Resuspension

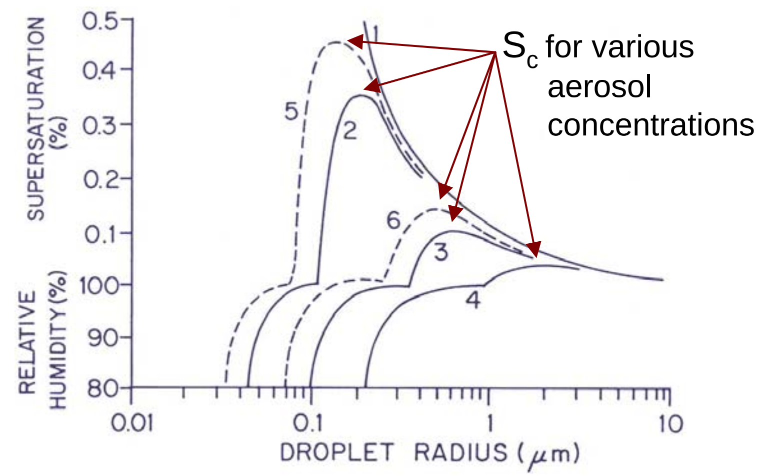
interstitial → cloud-borne → interstitial

activation ***resuspension***



aerosols activated when their critical supersaturation, S_c, is less than the environmental supersaturation at cloud base

Köhler Curves



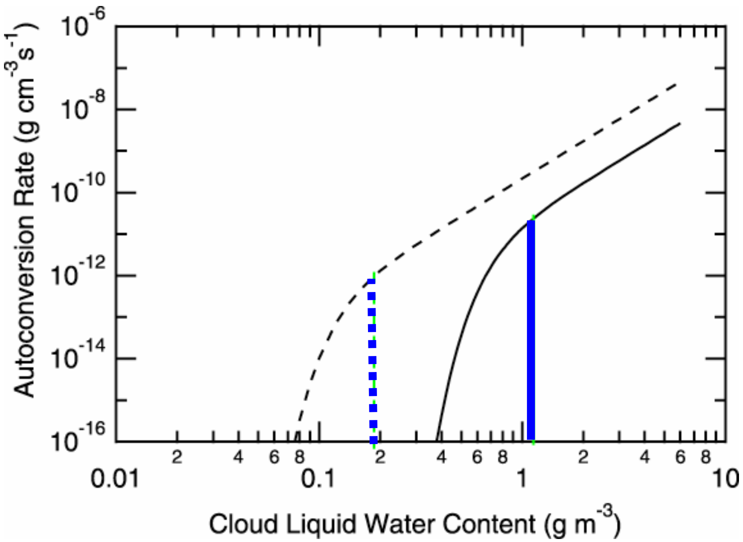
1: pure H_2O
2: 10^{-19} kg NaCl
3: 10^{-18} kg NaCl
4: 10^{-17} kg NaCl
5: 10^{-19} kg $(NH_4)SO_4$
6: 10^{-18} kg $(NH_4)SO_4$

S_c, depends on aerosol size and composition

Köhler curve plot: Fig. 4.12, Wallace and Hobbs, 1977

Autoconversion of Cloud Droplets

- autoconversion: coalescence of cloud droplets to form embryonic rain drops
- replaced autoconversion parameterization employed by *Lin et al.* microphysics scheme with *Liu et al.* [2005] parameterization
  - adds droplet number dependence
  - physically based w/o tunable parameters



Black: New Liu et al. parameterization
Blue: Kessler-type parameterization, similar to default Lin et al. scheme

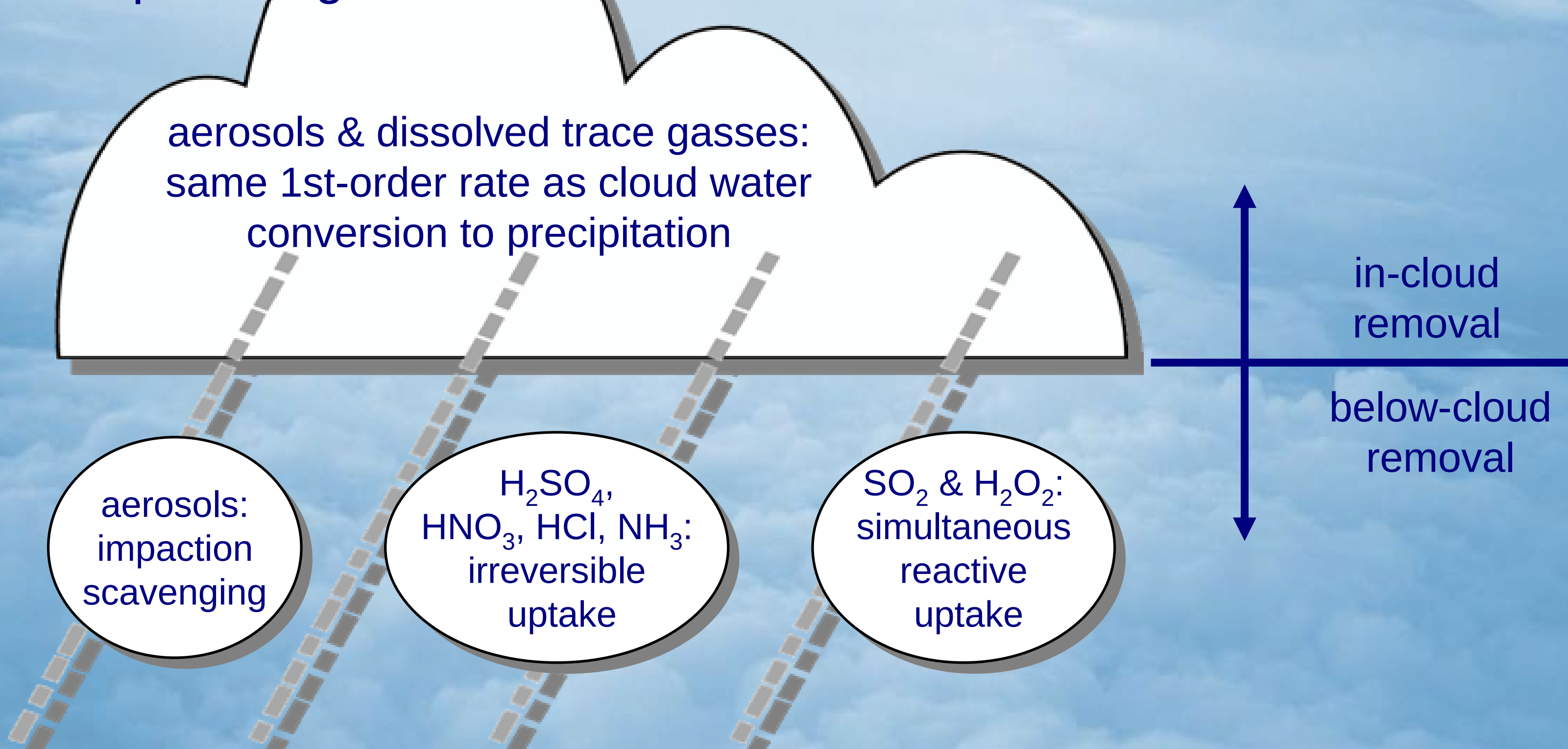
Dashed: N = 50 cm^{-3}
Solid: N = 500 cm^{-3}

Adapted from Fig. 3, Liu et al., 2005, GRL.

Aqueous Chemistry & Wet Removal

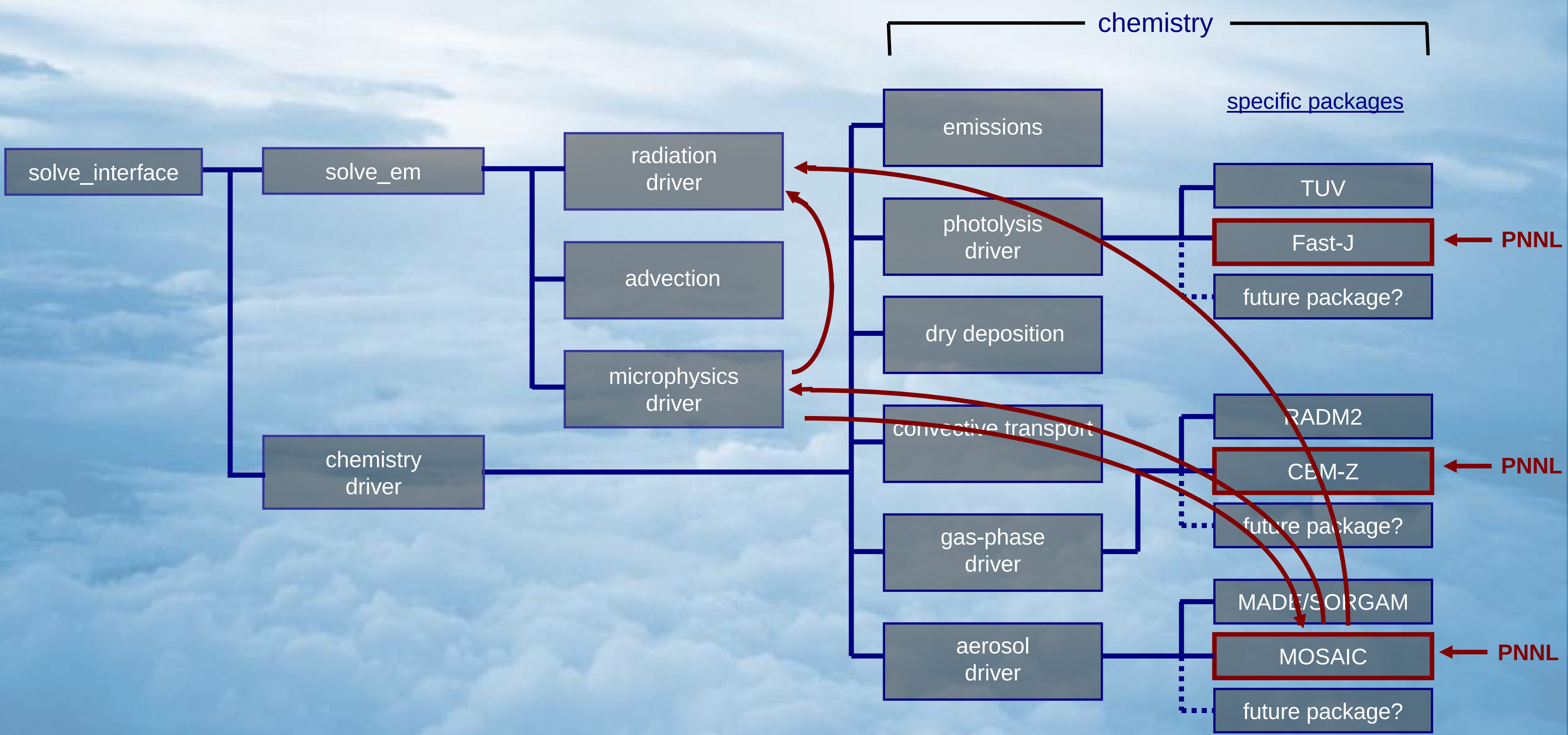
Aqueous chemistry:

- based on *Fahey and Pandis [2001]* and *Pandis and Seinfeld [1989]*
- leads to particle growth across size bins



scavenged aerosols and gases are assumed to be instantly removed (wet deposited)

WRF-Chem Flow Chart



1) **aerosol-radiation feedback: pass τ, ω_o, *g* at 300, 400, 600, 1000 nm into *module_ra_gsfcw.F***

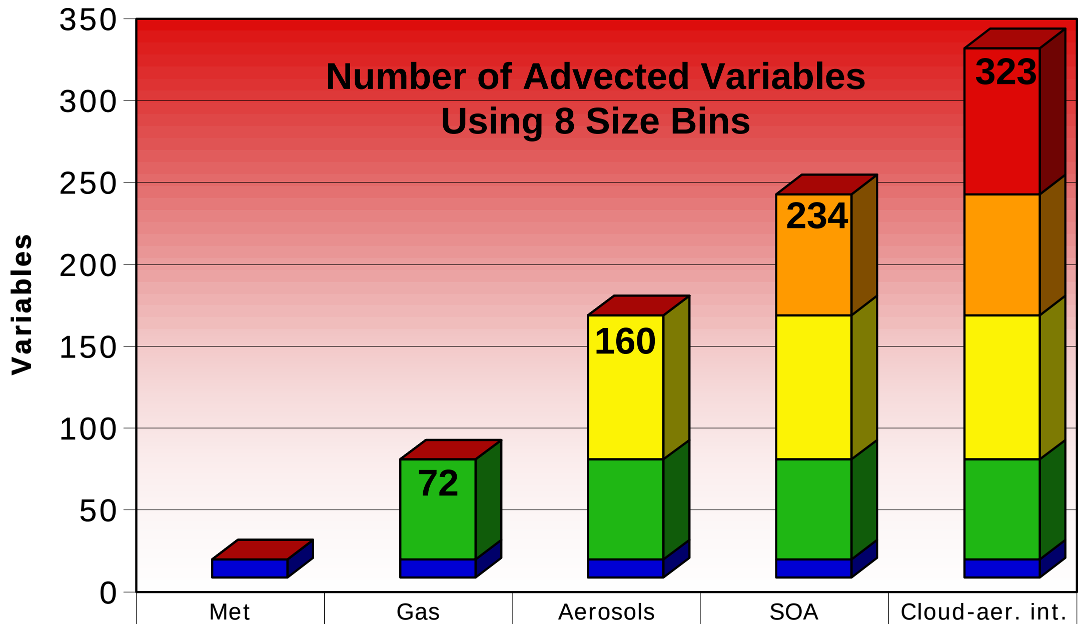
2) **cloud aerosol interactions: update *qndrop* and pass into *module_mp_lin.F***

3) **cloud aerosol interactions: transfer cloud properties to update cloud chemistry and scavenging**

4) **aerosol-cloud-radiation feedback: pass *qndrop* into *module_ra_gsfcw.F***

Computational Considerations

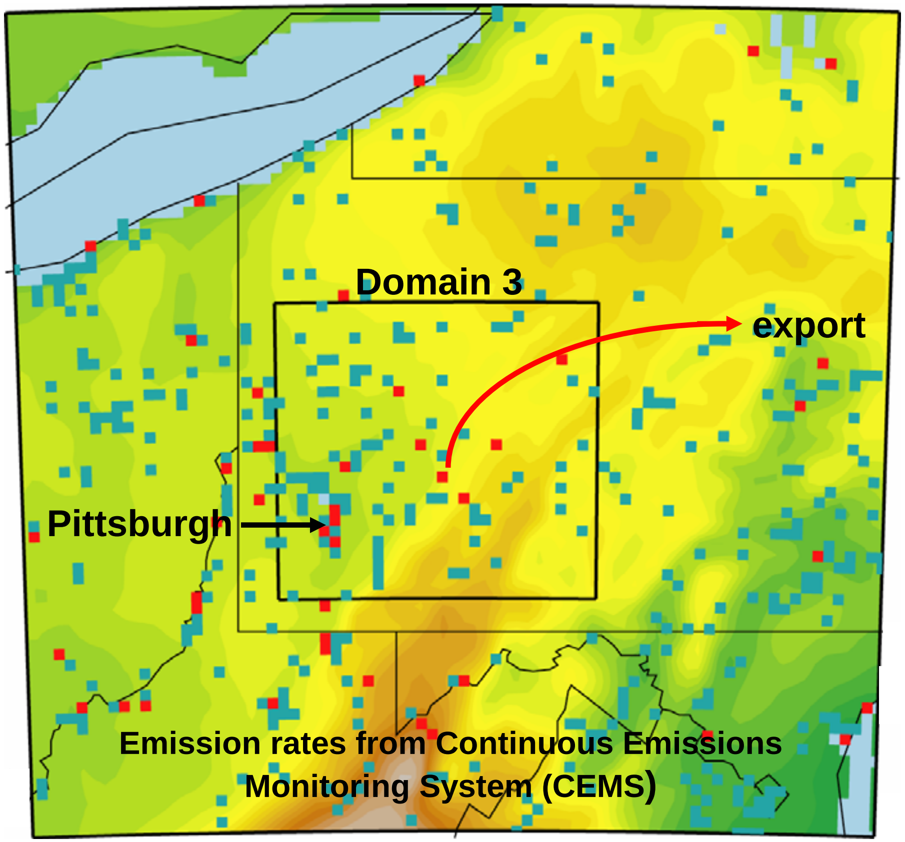
- chemistry, particularly the sectional aerosol approach, requires a very large number of variables.
  - computationally expensive
  - memory intensive



	Met	Gas	Aerosols	SOA	Cloud-aer. int.
Cloud-aer. int.	0	0	0	0	89
SOA	0	0	0	74	74
Aerosols	0	0	88	88	88
Gas	0	61	61	61	61
Met	11	11	11	11	11

Configuration

Case Study: Western Pennsylvannia

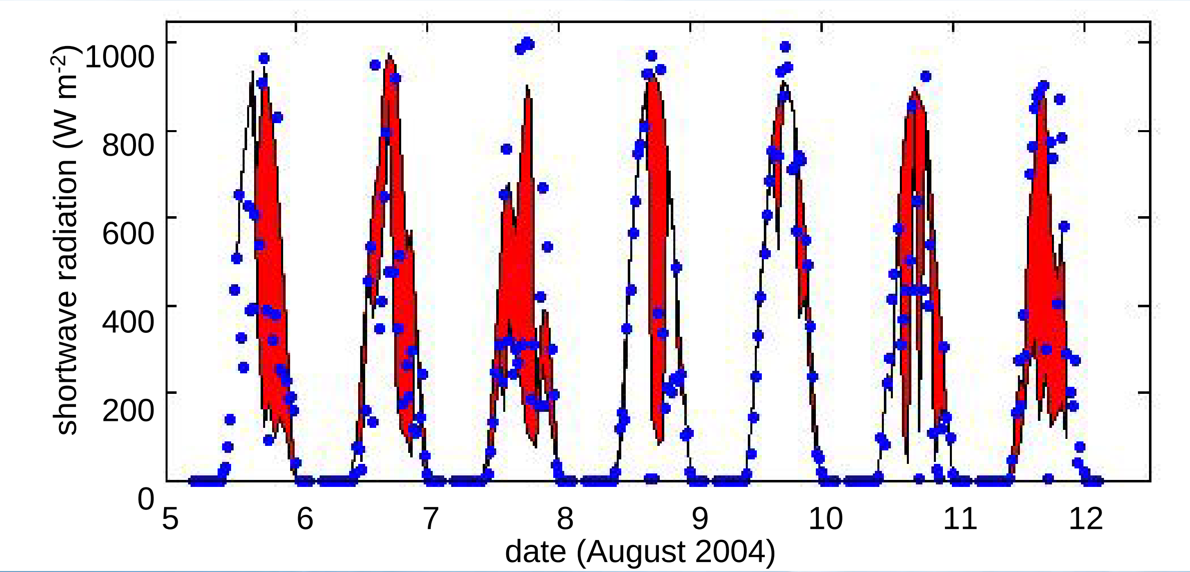


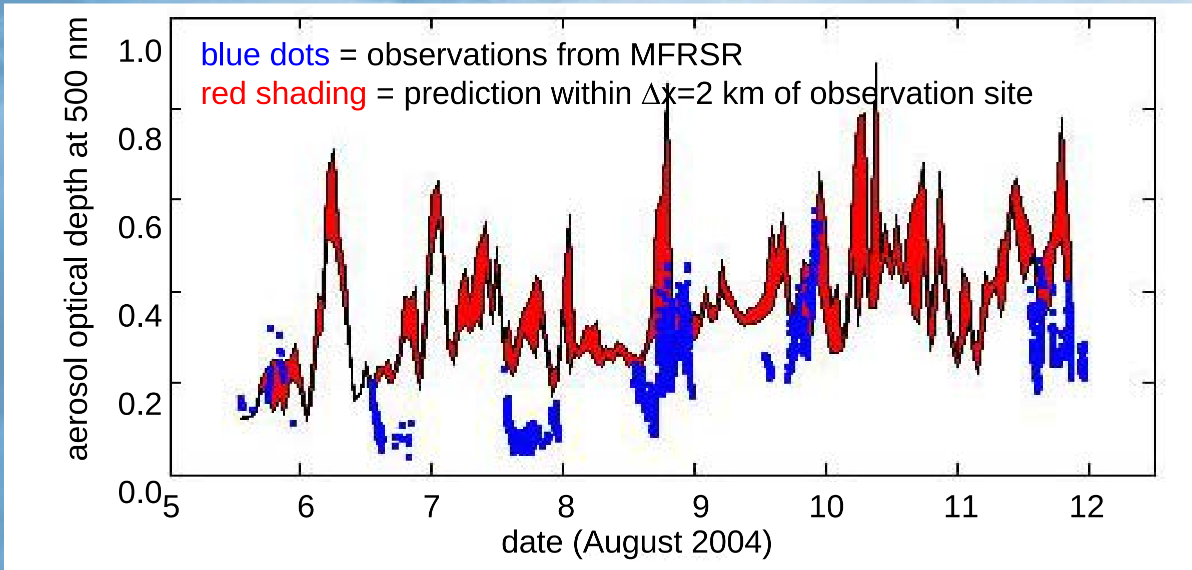
Domain	Δx	Δt_met	Δt_chem	nodes
1	18 km	90 sec	5 min	107x103x57
2	6 km	30 sec	5 min	82x79x57
3	2 km	10 sec	5 min	91x85x57

Objective: test new modules in WRF-chem for simulating cloud-chemistry and cloud-aerosol interactions downwind of major SO_2 point sources

- 5-11 August 2004
- periods of cloudy conditions and rain
- specifically examine
  - sulfate formation from SO_2
  - impact of clouds on sulfate
  - impact of aerosols on clouds
  - impact of midwest emissions on particulates along the east coast
- 5 and 3 times real time w and w/o cloud-aerosol interactions, respectively for 16-processors
- 1.3 and 0.9 Gb of memory w and w/o cloud-aerosol interactions, respectively

Surface Data



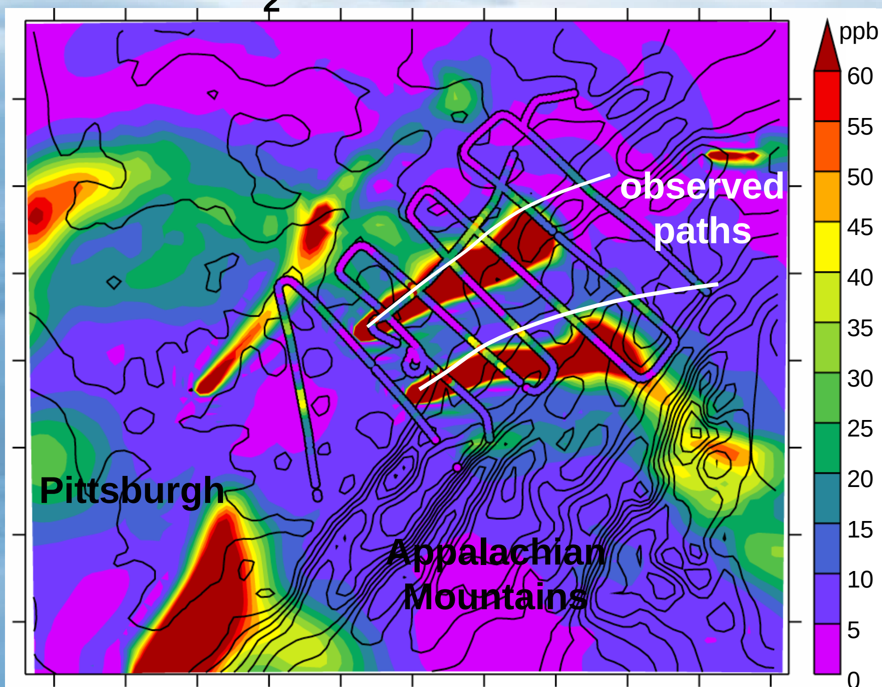


- only one day almost cloud free (9th)
- predicted reductions in SW radiation resulting from clouds similar to observed, although large differences at times
- predicted aerosol optical depth (AOD) close to observed on the 5th, 8th, and 9th and somewhat higher than observed on the 7th, 8th and 11th
- large spatial variations in AOD seen in horizontal plots

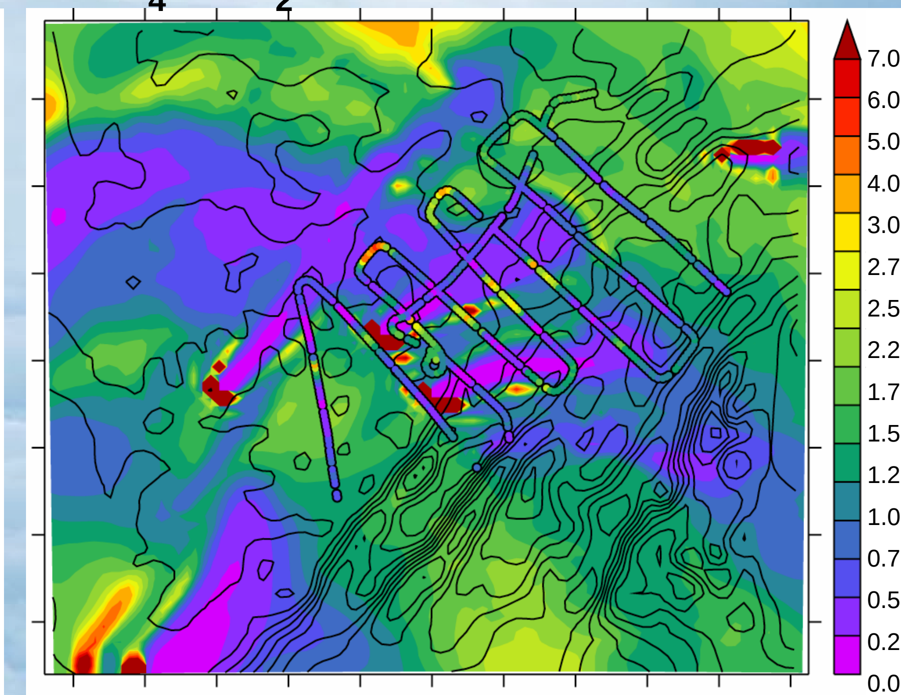
Power Plant Plumes

Predictions at 18 UTC 9 August 2004

SO_2 at ~900 m MSL



SO_4 / SO_2 Ratio at ~900 m MSL

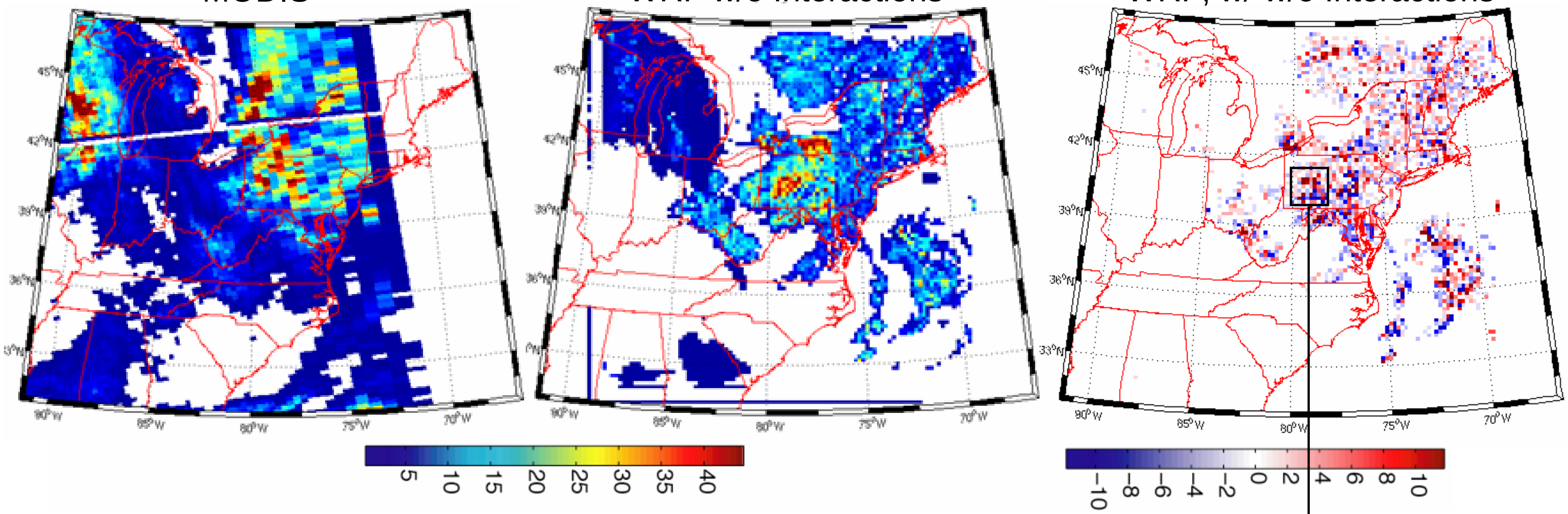


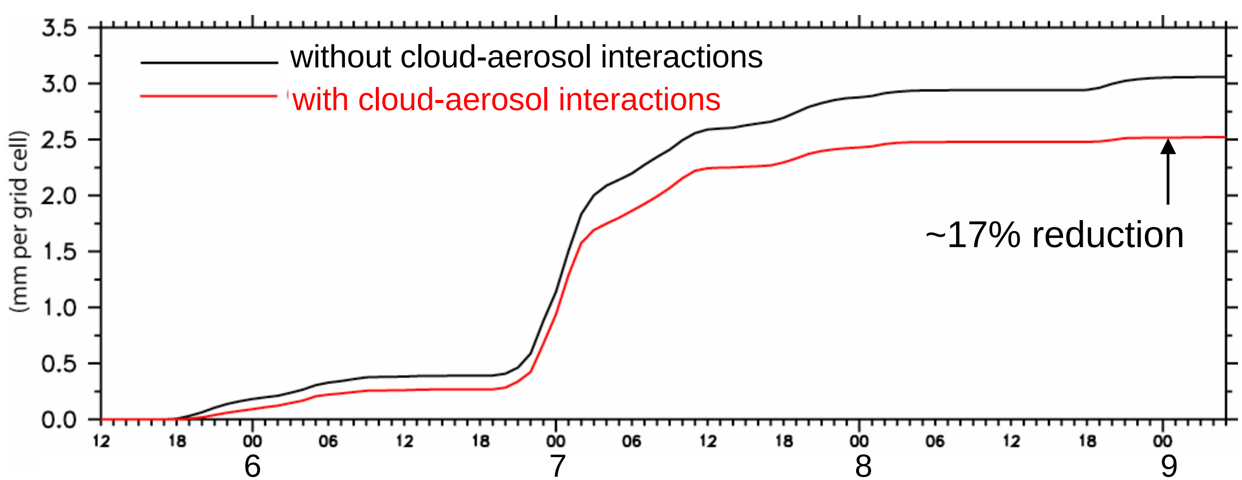
DOE G-1 Aircraft Observations 16:30 - 1930 UTC

- predicted wind directions more westerly than observed, resulting in small errors in predicted SO_2 plume paths
- SO_2 and SO_4 higher than observed, but SO_4 / SO_2 in good agreement with observations

Impacts on Meteorology

Cloud Optical Depth, 19 UTC 7 August 2004

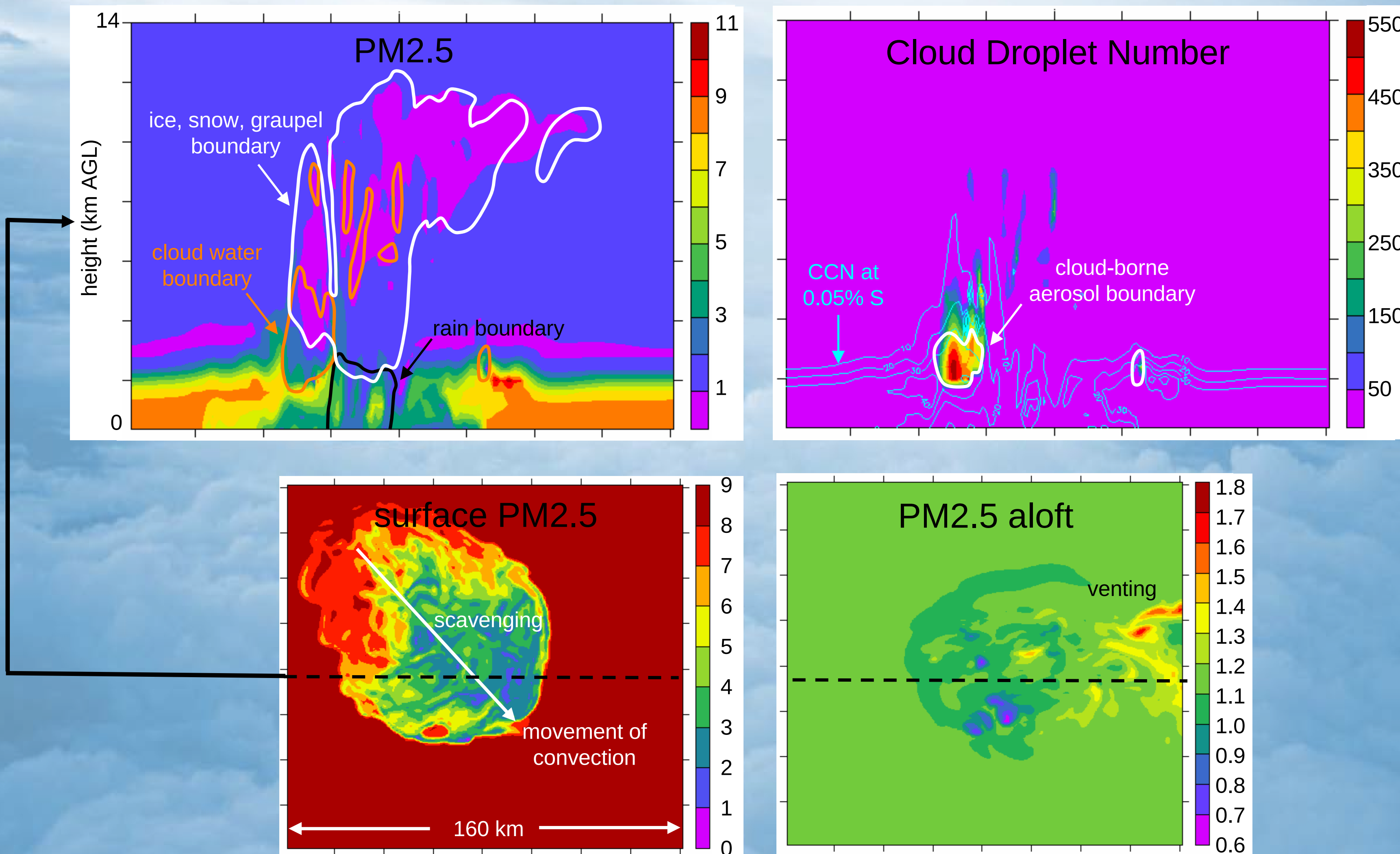




Domain 3 Grid Cell Average Precipitation Amount

Explicit Convective Cloud Simulation

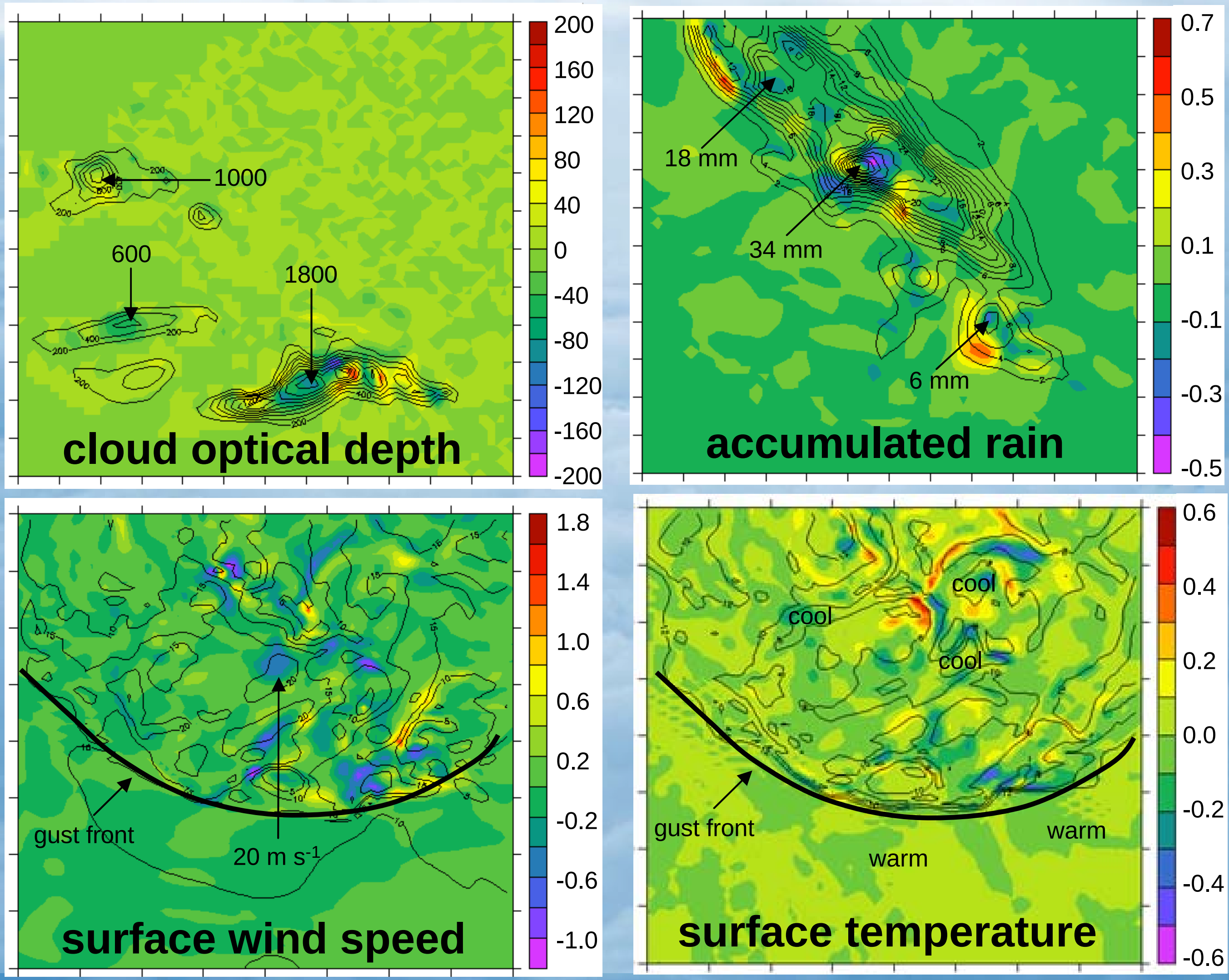
Results from Idealized Case (Δx = 1 km) at 1.5 h



most aerosols scavenged, but small fraction vented into free atmosphere

Effects of Boundary Layer Aerosol

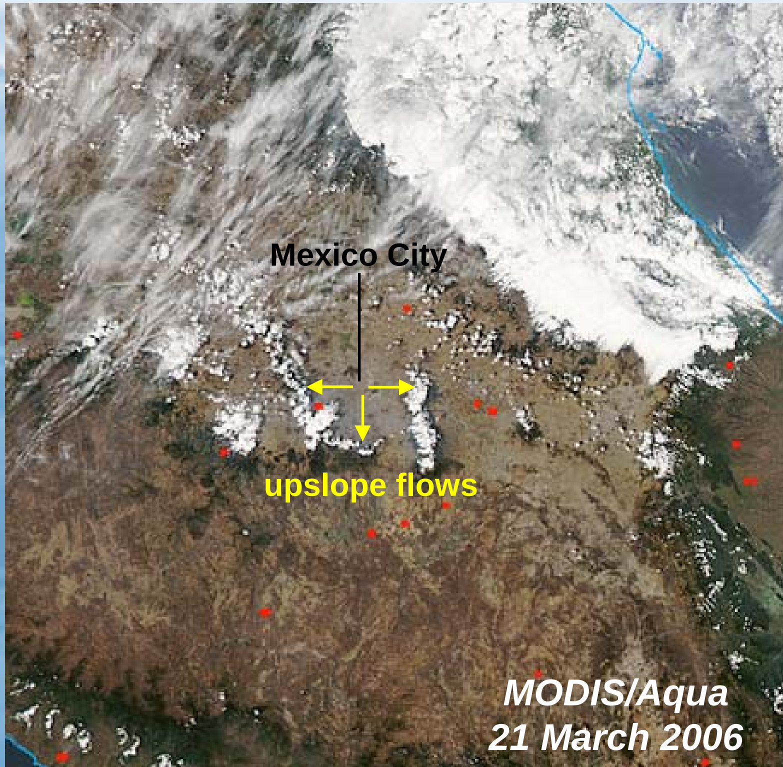
Black contours = BL 9 μg m^{-3} simulation
Color = Differences (BL 9 μg m^{-3} - BL 18 μg m^{-3} simulation)



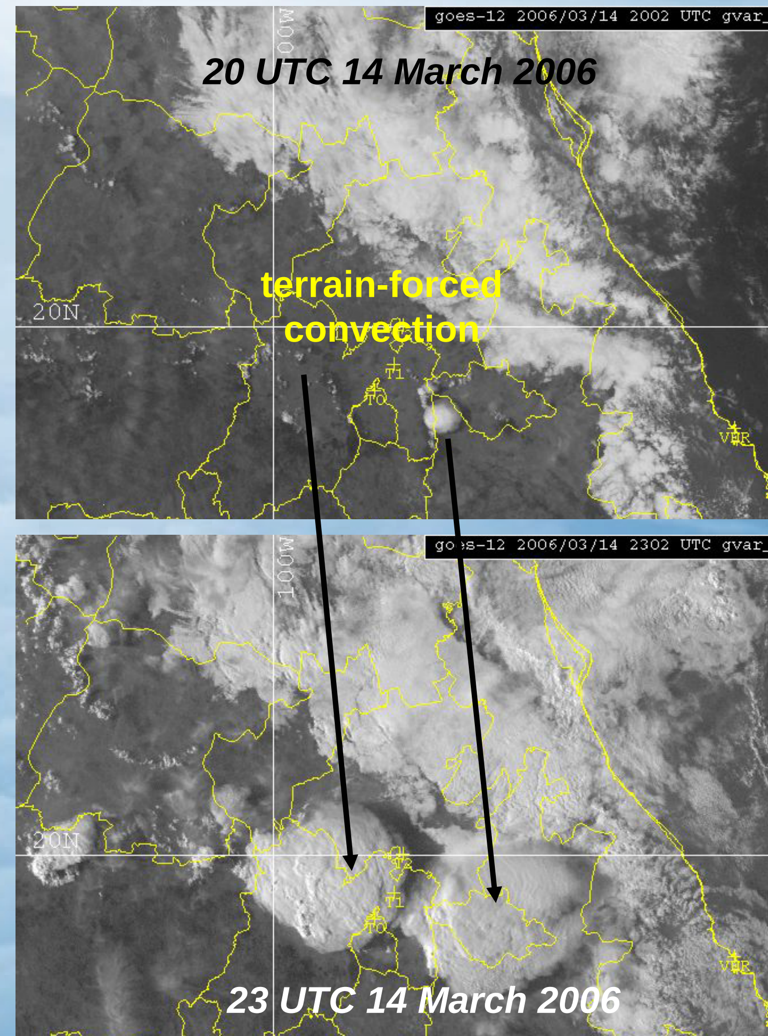
changed by as much as 5 mm over 3-h simulation

Clouds and Megacity Pollutants

Shallow Convection



Deep Convection



- particulates vented by convection into free troposphere and scavenged by precipitation
- to what extent to large anthropogenic particulate sources affect cloud properties?

Summary of WRF-chem Development

- cloud chemistry and cloud-aerosol interactions implemented this year
  - includes aerosol indirect effects by changing cloud albedo and precipitation
  - coupled to cloud water only - possible to extend to ice, snow, graupel
  - currently being coupled to *Thompson et al.* microphysics
  - PNNL modules available in repository "soon"

- can now use WRF-chem to examine local and regional climate issues and air quality - climate interactions
  - framework set up for regional climate model based on WRF, although current schemes may need to be simplified for long simulation periods
  - simulate local climate forcing, not just down-scaling from global model

- on-going model development:
  - use high-resolution cloud-aerosol simulations to develop parameterizations of vertical mixing of aerosols for global climate models (Ghan)
  - SOA, coagulation, and nucleation compatible with MOSAIC (Zaveri)